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Nuclear Safety Information Center, Oak Ridge National Laboratory

Reactor Technology
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TO THE READERS

We regret that economic necessities compel us to terminate *Isotopes and Radiation Technology* with this issue.

The AEC expresses its appreciation to the editors and staff members of the Isotopes Information Center, Oak Ridge National Laboratory, who have helped prepare this journal for the past 9 years. The journal has made a significant contribution to the progress of nuclear technology.

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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Commercial Radioisotope Production in Power Reactors

By Joseph E. Machurek* and E. Eugene Fowler*

If one were to set out deliberately to design a reactor *least* suited to commercial radioisotope production and marketing, the result would be a nuclear power reactor. It is true that, in the last 10 years, more than 60 papers and reports on radioisotope production in power reactors have appeared in the technical literature. However, the majority of these are addressed to the production of the transuranium isotopes, principally ^{237}Np, ^{238}Pu, and ^{242}Cm. A number of these publications review the projected availability and speculated future economic value of fission-product radioisotopes from power-reactor operation. Only a handful of reports are extant on power-reactor production of radioisotopes for which there are current, growing, and discernible future markets. This latter circumstance is not too surprising. The fact is that virtually all design features of modern power reactors which contribute to efficient, economical production of electric power are in direct opposition to the practical requirements of commercial production and marketing of all but a few radioisotopes.

In considering the possible role of nuclear power reactors in this area, it is necessary to look beyond their mere technical capabilities for radioisotope production to the practical demands of the marketplace. It then quickly becomes apparent that, of the approximately 100 radioisotopes in current use, not more than perhaps half a dozen lend themselves to production in nuclear power reactors. On the basis of the current status of radioisotope applications technology, only a few could be said, with even a minimum degree of confidence, to possess a demand potential that would be significant relative to nuclear-power-reactor production capacity. However, it must also be said that, assuming certain technological developments in radioisotope applications, the demand for these half-dozen or so isotopes could exceed by orders of magnitude the total of the current radioisotope market. In that event, they should offer highly attractive investment opportunities to nuclear-power-reactor operators.

It is not the purpose of this article to denigrate the utility and potential importance of nuclear power reactors as a source of radioisotopes. On the contrary, assuming the technological developments previously mentioned, power reactors will be the only source of particular radioisotopes in quantities adequate to meet market requirements. Moreover, the applications in which they would be used would be of critical importance, ranging from national defense to public health. The purposes of this article are constructive and are twofold: (1) to identify, and then suggest that we not fruitlessly dissipate our energies on, those areas of radioisotope production in which power reactors can serve no useful role; and (2) to focus attention on those isotopes which (*a*) are especially well suited to production in power reactors; (*b*) appear potentially to offer attractive investment opportunities to power-reactor operators; and (*c*) would serve important requirements of the public and national interest.

With respect to the first objective, several features of modern power reactors are inconsistent, from an

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overall standpoint, with the practical requirements of commercial radioisotope production and marketing. The most relevant of these features are the relatively low neutron flux, the annual refueling cycle, and normal operation with control rods out of the reactor. The relatively low neutron flux in itself eliminates from consideration a host of isotopes currently in routine use. Medical and biological applications of radioisotopes, for example, require the highest possible specific activity, which is achievable only in neutron fluxes ranging from around 10^{14} to as much as 10^{15} n/(cm^2)(sec).

The annual refueling cycle is a particular disadvantage because many of the radioisotopes in largest demand are short-half-life nuclides. In production they reach saturation activity in a relatively short time—substantially less than 1 year—but the refueling cycle precludes removal of the product from the reactor in less than a year. Also because of these short half-lives, many customers require frequent deliveries, often semiweekly, and the 1-year refueling cycle again precludes accommodation of this practical market requirement. These drawbacks could be overcome by penetrations into the reactor pressure vessel, but this is not particularly attractive from the standpoints of safety and economics.

The "out-of-reactor" control-rod feature is mentioned because control rods have been frequently cited as copious sources of little- or no-cost isotopes. The premise here is that control rods could be fabricated of a material or materials that would perform the functions required of control rods and concurrently result in the production of useful radioisotopes. The flaw in this approach is that in an efficiently operating power reactor the control rods will seldom be in the reactor. At best, therefore, control rods would be an unreliable and unpredictable source of radioisotopes.

SUITABLE ISOTOPES

Application of the cited reactor design and operating constraints to the total list of isotopes in current or anticipated use indicates only four that appear at this time to merit consideration for possible future production in nuclear power reactors. These are in addition to the fission-product radioisotopes that are necessarily produced in reactors and are discussed below. The four isotopes identified meet the following criterions, which are, in effect, corollaries to the previously discussed constraints: (1) They are relatively long lived; (2) they can be efficiently produced by long irradiations at a low neutron flux; and (3) their production capacity in power reactors is not grossly disproportionate to reasonably anticipated future market demand, or, at least, such demand is significant enough to make them appear attractive for production. It is axiomatic, of course, that production of the isotopes cannot affect reactor operations adversely. Displacement of fuel elements, for example, to permit insertion of radioisotope targets would not seem to be very practical. On the other hand, placement of targets in unused rod-cluster control assembly locations in pressurized-water reactors (PWRs) or in vacated control-curtain locations in boiling-water reactors (BWRs) would appear to be reasonable.

On these bases, the radioisotopes other than fission products which merit consideration for possible future production in nuclear power reactors are ^{238}Pu, ^{3}H, ^{14}C, and ^{60}Co. Other isotopes—e.g., ^{244}Cm, ^{210}Po, and ^{170}Tm—would have been included in this list were it not for the fact that at present there is no basis for estimating future market requirements. All these isotopes have potential utility, but no significant specific applications have yet materialized. Should this situation change, these isotopes would clearly merit reconsideration for production in nuclear power reactors.

Each of the selected isotopes will now be briefly discussed in terms of best judgment as to estimated future requirements and power-reactor production capacity. The latter will be expressed as kilograms or kilocuries per year per 1000 MW(e) of installed power capacity in a PWR. For a BWR the radioisotope production capacity would be about 60% of that of a PWR. Additionally, production of any one isotope at maximum capacity would preclude concurrent production of any other isotope. It must be noted also that an increased fuel enrichment is required to compensate for the reactivity loss due to the presence of the target and to produce the same burnup. This will vary with the type and quantity of the target, but generally will be of the order of a 1% additional enrichment.

For ^{238}Pu, best judgment is that a total of 1 thousand kg will be required over the 10 years through 1980, with the rate of demand being about 3 hundred kg/year at the end of this period. Plutonium-238 production capacity in a PWR from irradiation of ^{237}Np is estimated to be between 31 and 82 kg/year per 1000 MW(e) power capacity, the range representing various target loadings.

U.S. Atomic Energy Commission (AEC) commercial sales of tritium are about 350 kCi/year and over the last several years have been increasing at an average rate of 20% annually. Assuming that this trend

continues, the total requirement over the 10 years through 1980 would be 8 thousand kCi, with the rate of demand being about 15 hundred kCi/year at the end of this period. The maximum production capacity for tritium in a PWR is about 150 thousand kCi annually per 1000 MW(e). This, of course, grossly exceeds the possible market demand. The latter nonetheless is significant and could be accommodated by power reactors using less than maximum target loadings.

The demand for ^{14}C in the United States is about 0.2 kCi/year and has been increasing about 10% annually. On this basis, the total requirement over the 10 years through 1980 would be 3 kCi, with the rate of demand being about 0.5 kCi/year at the end of this period. The maximum power-reactor production capacity for ^{14}C is about 2.8 kCi/year per 1000 MW(e) in a PWR. Here again, the market could be accommodated by less than maximum target loadings.

Cobalt-60 is a particularly interesting case in that it is already being commercially produced in limited quantities in a power reactor. About 2 thousand kCi/year is being manufactured in the Big Rock Point Nuclear Power Plant. Moreover, Canada is planning to produce the isotope in its Pickering reactors at a rate by 1975 of about 15 thousand kCi/year and, additionally, is loading cobalt targets into its Nuclear Power Demonstration Reactor. A recent market evaluation conducted for the AEC indicates a total demand for ^{60}Co over the 10 years through 1980 of about 90 thousand kCi, with a rate of demand at the end of the period of about 16 thousand kCi/year. These estimates are based on further extension of existing technology and do not assume any technological breakthroughs. The 1980 demand rate estimate must be treated with particular caution, however, since at about that time power-reactor-produced ^{137}Cs might become available for use, as discussed below. This isotope could encroach to some extent, and perhaps significantly, on the demand for ^{60}Co. Maximum power-reactor-production capacity for ^{60}Co in a PWR lies between 9 thousand and 30 thousand kCi/year per 1000 MW(e), the range reflecting various target loadings.

As indicated above, only one isotope can be produced at the maximum production capacities cited. As a practical matter, one would aim for a product mix most responsive to market demand, with no one isotope likely to be produced at maximum capacity.

Fission Products

Let us turn our attention now to the fission products generated from power-reactor operation—principally ^{137}Cs, ^{90}Sr, and ^{147}Pm. All these nuclides have current or developing applications that could result in significant future market demands. The nuclear fuel reprocessors, of course, would be in the best position from a practical standpoint to undertake chemical separation of these isotopes from the fission-product wastes. However, the potential markets for these products also might prove to be of interest to power-reactor operators. It should be borne in mind that, because of the indicated market-demand growth rates, the establishment of a private fission-products separation and distribution industry would be unlikely before about 1980.

Power-reactor-produced ^{137}Cs is unique in that it contains large quantities of ^{134}Cs. For technical reasons, the presence of the ^{134}Cs significantly enhances the market value of power-reactor-produced ^{137}Cs as compared to the radioactive cesium product currently available from AEC production reactors. An AEC-sponsored market evaluation of production-reactor-produced ^{137}Cs indicates a total demand over the 10 years through 1980 of about 170 thousand kCi, with the rate of demand at the end of the period being about 30 thousand kCi/year. For the power-reactor-produced ^{134}Cs–^{137}Cs mixture, these estimates would be equivalent to a total demand of 126 thousand kCi and a demand rate in 1980 of 24 thousand kCi/year, stated in terms of the ^{137}Cs content only. As already alluded to, the ^{134}Cs–^{137}Cs mixture and ^{60}Co will compete for the same markets. The 1980 estimated demand rates therefore are subject to modification depending on the relative share of the market each gains. Their respective market shares will be determined almost solely by their prices relative to each other. Cumulative availability of the ^{134}Cs–^{137}Cs mixture from power-reactor operations over the 5 years through 1980 is estimated at 660 thousand kCi (in terms of ^{137}Cs), with the material being generated in 1980 at a rate of 212 thousand kCi/year. The 5-year period was selected as being the maximum time under AEC regulations that fission-product wastes could be retained in liquid form for subsequent separation of the ^{137}Cs product, assuming advent of a private separation capability in 1980.

Demand for ^{90}Sr over the 10 years through 1980 is estimated to total 68 thousand kCi, with a demand rate at the end of the period of about 12 thousand kCi/year. The credibility of these estimates is enhanced by a recent request from the Department of the Navy to the AEC for an availability and cost quote on 20 thousand kCi of ^{90}Sr. Cumulative availability of ^{90}Sr from power-reactor operations over the 5 years

through 1980 is estimated at 475 thousand kCi, with the material being generated in 1980 at a rate of 150 thousand kCi/year.

Market-evaluation data currently available on ^{147}Pm cover only the 5-year period through 1974, the total demand being estimated at 58 hundred kCi and a demand rate in 1974 of 28 hundred kCi/year. The cumulative availability of ^{147}Pm over the 5 years through 1980 is estimated at 255 thousand kCi, with the material being generated in 1980 at a rate of 120 thousand kCi/year. It is important to note that, because ^{147}Pm has a half-life of only 2.6 years, much of it is lost through decay during storage.

The practical feasibility of nuclear fuel reprocessors engaging in commercial production of separated fission products obviously is dependent on their ability to obtain an adequate return on investment through sale of the materials at prices compatible with market requirements. This has been studied in very great detail. All cost and price figures cited in the following discussion are in terms of 1971 dollars. The study was predicated on a 10-year economic life for the required facilities and a 15% after-tax return on investment. These ground rules were adopted after discussions with the current and prospective nuclear-fuel processors. It was further established that the market values for power-reactor produced ^{134}Cs–^{137}Cs mixture and ^{90}Sr were 30 and 10 cents/Ci, respectively. The figure for ^{134}Cs–^{137}Cs was arrived at by calculating its value relative to ^{60}Co, currently being marketed at around 40 cents/Ci. Although this price possibly could slip in the future, it was judged that in a mature market, such as that assumed for the 1980s for the purposes of this study, any price erosion would be modest. The ^{90}Sr value was derived from an evaluation of the economic requirements of the applications in which it would be used. A detailed analysis was made of variously assumed market demands, plant capacities, and plant startup dates, together with associated capital costs in each case, to determine if there could be found a combination of these parameters which approximated the conditions that would be required to induce the nuclear-fuel processors to enter the market. An analysis of 29 cases indicated that a plant starting up in 1982, having a capacity for 27 thousand kCi/year of ^{134}Cs–^{137}Cs mixture and 12 thousand kCi/year of ^{90}Sr, marketing these products at levelized prices of 20 and 10 cents/Ci, respectively, and having been erected at a capital cost of $17.3 million, would provide a 15% after-tax return on investment based on a 10-year economic life. As will be immediately evident, the plant capacity almost exactly matches the estimated market demand in 1980 for the ^{134}Cs–^{137}Cs mixture and ^{90}Sr as previously identified. Perhaps it would be well to state at this point that the market evaluations and plant studies were done by separate staffs completely independent of each other. No special analysis was made of ^{147}Pm production since it appeared that this could be readily accommodated in a viable cesium–strontium plant. These analyses suggest that, taking into account all relevant factors, private separation and marketing of fission products from nuclear power reactors in the early 1980s may well be within the realm of economic feasibility.

. . .

It is customary to end a presentation such as this with some scholarly recital of conclusions and recommendations. It would appear the better part of valor in this case, however, to be content with having presented the situation as we see it, leaving the final evaluation and conclusions to those whose money will be "on-the-line."

(MG)

CORRECTION

On page 158 of Vol. 9, No. 2, of this journal, Table 6 of the ^{123}I article by Sodd et al. should read as follows:

Table 6 Yields from the ^{122}Te(^{3}He,2n)^{123}Xe and ^{122}Te(^{3}He,pn) Reactions

Energy, MeV	Target wt., mg/0.7 cm^2	Yield			
		^{123}Xe, μCi/μA-hr	^{124}I, %	^{123}I, μCi/μA-hr	Total ^{123}I, μCi/μA-hr
		80.4% ^{122}Te Target			
14.7 ± 0.9	9.8		0.63	23	
14.2 ± 1.4	13.8		0.83	18	
		73.3% ^{122}Te Target			
13.5 ± 0.8	7.8	0.18		0.06	0.07
17.9 ± 0.6	5.4	13.7		6.3	6.7

Section II

Isotopes and Radiation Technology

Isotope Technology Development

Neutron Activation Analysis in Scientific Crime Investigation

By Vincent P. Guinn*

Supplementary Keywords: *forensic investigation; review, technical.*

Abstract: *The literature (91 references) is reviewed on applications of neutron activation analysis in forensic investigations.*

Because of the great sensitivity of high-flux (reactor) thermal-neutron activation analysis (NAA) for the detection and quantitative determination of a large number of elements, the method has been applied in scientific investigation of crime. The literature on this subject is voluminous (e.g., Refs. 1 and 2 with 135 and 168 titles, respectively). Often, specimens can be analyzed which are too small for meaningful analysis by more conventional methods, and trace constituents can be determined to indicate the possibility of common origin. The nondestructive nature of the instrumental form of the method is a particular advantage in forensic work since the samples are not destroyed during analysis but can be subsequently presented, intact, in court.

The forensic NAA research and development work has, to date, been conducted chiefly at Gulf General Atomic in San Diego, Calif., and Oak Ridge National Laboratory in Oak Ridge, Tenn., in the United States; at the Centre of Forensic Sciences and the University of Toronto in Canada; and at the Home Office Laboratory in Aldermaston, England. However, significant contributions have been made by groups in Scotland, the Netherlands, Brazil, Japan, and other countries. The technique is being used regularly for investigating criminal cases at: Gulf Energy & Environmental Systems in San Diego; the U. S. Treasury Department and the Federal Bureau of Investigation Laboratories in Washington, D. C.; the Centre of Forensic Sciences in Toronto; and the Home Office Laboratory in Aldermaston. Results of NAA were first presented in a U. S. court in March 1964, and there have since been more than a hundred such presentations in the United States, a number in Canada and England, and some in other countries. A 28-min color and sound film[3] prepared by Gulf General Atomic for the U. S. Atomic Energy Commission—"The Nuclear Witness: Activation Analysis in Crime Investigation"—is based largely on actual cases investigated and is used by law-enforcement agencies for training agents.

In 1966, the First International Conference[4] on Forensic Activation Analysis was held in San Diego, cosponsored by Gulf General Atomic, the U. S. Atomic Energy Commission, and the California Association of Criminalists. Immediately after this conference, the speakers met and formed an informal international Forensic Activation Analysis Coordinating Group, whose purpose was to aid in the proper and systematic development of this field and of its use in court in criminal cases. Membership is open to all persons actively engaged in forensic activation-analysis studies. The second such conference is scheduled to be held in Glasgow, Scotland, Sept. 27–29, 1972.† More re-

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cently, the International Atomic Energy Agency (IAEA) has initiated a program in forensic activation analysis. In May 1969, the Agency held a consultant's meeting in Vienna to formulate recommendations for Agency activities in this field. As an outgrowth of this meeting, the IAEA plans to initiate several activities in this field—e.g., preparation of a periodically updated bibliography and preparation and issuance of a state-of-the-art paper on forensic activation analysis. Along with Ref. 1, the U. S. National Bureau of Standards' bibliography will be incorporated in the IAEA bibliography. Actual case applications of activation analysis and aspects of the presentation of NAA results in court are reported in Refs. 4*a*,*b*, 5, 6, 7*a*, and 8–10. Reviews of forensic activation analysis are found in Refs. 4*c*, 7*b*, 11*a*, 12–17, 18*a*, 19, and 20.

GUNSHOT-RESIDUE STUDIES

In late 1961, Guinn and coworkers at Gulf General Atomic, in cooperation with R. H. Pinker, Chief Criminalist of the Los Angeles Police Department, obtained promising results in exploring the possibility of detecting traces of barium and antimony on the back of the gunhand of a person who had recently fired a gun. They used NAA followed by carrier radiochemical separation of the induced 82.9-min ^{139}Ba ($E_\gamma = 0.166$ MeV) and 2.80-day ^{122}Sb ($E_\gamma = 0.564$ MeV). Most cartridge primers contain barium nitrate, Sb_2S_3, and lead styphnate, and some of the older ones also contain mercury fulminate. Since barium nitrate and Sb_2S_3 are more widely used (particularly in U. S. cartridges) and the NAA sensitivity for lead is not very good, attention was focused on the detection and quantitative determination of barium and antimony removed from a selected area of the back of the hand via a paraffin "handlift" procedure.

A more detailed study of gunshot residues was initiated in May 1962 with the support of the DID, additional support being provided, starting in 1967, by the U. S. Department of Justice. Variables that have been investigated include type, make, condition, caliber, cleanliness, barrel length, number of firings, and angle of firing of the gun; brand of ammunition; wind direction and velocity; spatial distribution of residues on the hand; methods of removal of the residues; and occupational "handblank" levels. Both labeled gunpowder and labeled primers have been used.[21-23] A comprehensive summary of this gunshot-residue work has been published.[24]

Several hundred special gunshot-residue paraffin hand-sampling kits[25] have been distributed to a large number of law-enforcement agencies in the United States and abroad during the past several years. The kits are frequently used by these agencies for examining the hands of suspects and apparent or questionable suicide victims in gunshot cases. The results of NAA determination of barium and antimony in the paraffin have been successfully presented in courts in both the United States and abroad. In an interesting test of the method in March 1970, six paraffin handlift samples (both hands of three persons) were prepared by the Dallas, Tex., Medical Examiner's Office and sent as unknowns to the Gulf Energy & Environmental Systems laboratory. The results of barium and antimony determinations by NAA were interpreted as indicating that two samples were from the hands of a person who had apparently not fired a handgun recently (or had thoroughly washed his hands after firing one); two were from the hands of a person who had recently fired a medium-output gun (such as a 0.38 caliber revolver) with his right hand; and two were from the hands of a person who had fired a higher output gun (such as a 9-mm, 0.45- or 0.25-caliber automatic), holding the gun with both hands. These results were the exact situation.

More recently, Scott, Coleman, and Cripps[26] performed autoradiographic measurements on neutron-activated paraffin handlifts and demonstrated the particulate nature of the deposits via 2.80-day ^{122}Sb. Subsequently, similar autoradiographic results were obtained at Gulf Energy & Environmental Systems by H. L. Schlesinger. The Ba–Sb technique has also been investigated in Japan.[27] In the United States, the earlier work has been extended[28,29] using rare earth–labeled gunpowder.

In closely related studies, NAA measurements of traces of antimony on surfaces such as skin and cloth have been used to estimate firing distances. This technique, developed in Germany,[30] has been considerably extended in Canada[4*d*,31] and used in the investigation of Canadian gunshot cases. Comparison of an evidence sample with test samples obtained by firing the recovered gun (rifle or handgun) with the same brand of ammunition, to yield an antimony concentration-vs.-firing distance calibration curve, showed that the firing distance can often be determined with reasonable accuracy up to distances of 8 to 10 ft.

The NAA method has also been used by Schlesinger, Hoffman, and Pro,[4*e*] by Krishnan,[31*b*] and by Krishnan and Nichol[32] to establish whether holes in various materials (tissue, cloth, leather, etc.) are or are

not bullet holes. Material around the suspect hole or swabbings of the area are activated and then examined for the presence of significant levels of ^{139}Ba and ^{122}Sb (and also for ^{64}Cu if a copper-jacketed bullet is involved).

TRACE-ELEMENT CHARACTERIZATION APPLICATIONS

In criminal investigations it is often necessary to establish, as definitively as possible, whether specimens of a particular type involved in a case do or do not have a common origin. Multielement high-flux instrumental neutron activation analysis has proved to be a powerful technique for such applications. In general, specimens of a given type of material (e.g., blue paint) will be very similar in their levels of major, minor, and trace elements if they are from the same specific source (e.g., blue paint of a particular brand, made by one particular manufacturer, and perhaps from a particular production batch). On the other hand, specimens of a given type of material but of different origin may or may not be fairly similar in their levels of major and minor elements and almost certainly will differ considerably in their levels of various trace elements.

The NAA trace-element characterization technique has been applied, both in extensive investigations and exploratory studies, to natural products (e.g., hair, opium, marijuana, and soil) and to manufactured products (e.g., paint, glass, bullet lead, and paper). The most extensive such investigations have been made on human head hair in Canada[4f,g,11b,33] and in England.[4h] In washed head-hair specimens[33] from a thousand persons (mostly Canadians), 20 elements were determined, although usually, under the conditions used, only 10 to 14 elements are found. The conditions were a 50-hr irradiation of a 5- to 10-mg sample with a thermal-neutron flux of 10^{13} n/(cm^2)(sec), followed by NaI(Tl) gamma spectrometry at several decay times. In the English work, specimens of head hair from 750 persons in England and Wales were analyzed similarly. The results showed element concentrations differing in some cases by as much as 3 to 5 orders of magnitude in samples from one person to the next but a maximum of about twofold in larger samples from any given person. Individual strands of hair from any one person showed greater variations than did the samples—an extreme of about 10-fold. In all, instrumental NAA of human hair has shown the presence of a total of 29 elements: Na, Al, Cl, K, Ca, Sc, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Mo, Ag, Sb, I, Cs, La, Sm, Eu, Au, and Hg. A number of papers have been concerned with the NAA determination of arsenic in human hair—usually with radiochemical separation—in connection with cases of chronic arsenic poisoning.[4a,i,7c,34-38] Numerous complicating factors and the proper interpretation of NAA results in comparing hair are discussed elsewhere.[4c,f,j,18b,39-41]

The instrumental NAA trace-element characterization technique has been explored on a large scale not only with hair but also with such evidence materials as paint,[4k,l,22,42-45] bullet lead,[21,22,46-48] tape,[49] and paper.[22,50] Small-scale studies have been made on drugs (opium, marijuana, heroin, and other dangerous drugs),[18c,22,51-56] glass,[4m,5,22] oils and greases,[7d,22,57] whiskey,[58,59] soil,[7e,22] and miscellaneous materials (rubber, wood, plastics, inks).[22]

(MG)

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A NEW METHOD OF ELEMENT ANALYSIS USING NUCLEAR RESONANCE SCATTERING OF GAMMA RAYS*

If a sample is irradiated with gamma rays of energy very slightly greater than the energy of the nuclear level being excited in an element being determined, detection of the resonantly scattered radiation gives a count rate proportional to the concentration of that element. The technique is simpler and more sensitive for a specific element than other nuclear methods now in use. Radioisotopes are available to initiate resonant gamma-ray scattering in at least 31 stable isotopes—e.g., ^{7}Be for Li, ^{75}Se for As, ^{183}Re or ^{184}Re for W, and ^{203}Hg for Tl.

The method was tested by determining copper in samples of known concentration in a simulated borehole—an 11.4-cm-diameter aluminum tube surrounded by concrete, with the sample inside the tube. The source used was gaseous $^{65}ZnI_2$. The sealed source was placed in a furnace inside the borehole, and pulses picked up by a NaI(Tl) crystal, shielded to prevent its detecting direct radiation, were fed through the usual electronics to a multichannel analyzer. The source strengths used were small (6.6 to 29 mCi), but, in field application, sources as large as 100 Ci might be used. The results showed a linear variation in the count rate with weight percentage of copper (Fig. 1).

The method was also tested for nickel determinations, using a 7-mCi $^{60}CoBr_2$ source.

The method could also be used for specific element determination in remote areas such as the ocean floor. (MG)

*Abstract of article by B. D. Sowerby, *Nuclear Instruments and Methods*, **94**: 45-51 (1971).

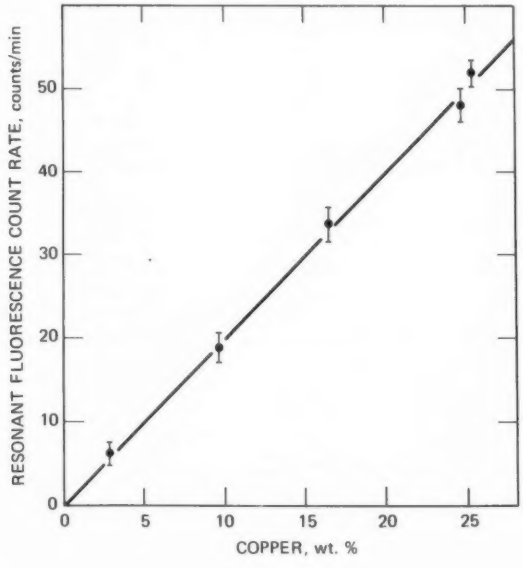

Fig. 1 Variation in resonance count rate with copper percentage.

A PORTABLE RADIOISOTOPE X-RAY FLUORESCENCE ANALYZER AND ITS APPLICATIONS*

Supplementary Keywords: *source, X ray; chemistry, analytical; metallurgy; instrument; spectrometry; theory, applied; differentiation;* ^{3}H; ^{55}Fe; ^{109}Cd; ^{147}Pm; ^{238}Pu; *Japan.*

A new portable X-ray fluorescence analyzer is described which consists of a probe (1.7 kg) and an electronic unit (7 kg). The radioisotopes ^{3}H, ^{55}Fe, ^{109}Cd, ^{147}Pm, and ^{238}Pu are used as X-ray sources. X-ray spectral analysis is done with a proportional counter and single-channel pulse-height analyzer. X-ray filters are used, if needed, and the characteristic X rays from the elements being determined can be selected with a rotary switch.

The performance of the instrument was demonstrated by analysis of alloys, minerals, and other materials. Detection limits of ~10 ppM were achieved in determinations of copper in an aluminum alloy and of Mo, V, W, and Zr in alloy steel. In differentiation of alloys according to the contents of these elements, a 0.2% difference can be detected in only 1 to 3 sec; to detect 1% difference in chromium and nickel contents of alloy steel, a slightly longer time—4 to 5 sec—is required. To determine elements of concentration >1% in minerals, a linear calibration curve without matrix correction can be used. The relative errors of measurements for elements of mean concentration of 1% and >15% are 10% and <3%, respectively. Detection limits for manganese are 10 ppM in aqueous solution and 2.2 μg/cm² deposited on a filter paper. (HPR)

*Summary of article (in Japanese) by T. Tanemura, H. Suita, and H. Yamamoto, *Radioisotopes*, **20**(9): 438-444 (1971).

Nuclear Techniques for Product Inspection and Quality Assurance*

By Robert F. Hill†

Supplementary Keywords: *industry, transportation; material tracing; radiography; measurement, moisture;* ^{82}Br; ^{241}Am; ^{75}Se.

Abstract: *The increasing emphasis on automotive safety and reliability through product assurance has stimulated the development of new and improved inspection methods. Nuclear techniques are finding ever-increasing application and have been used successfully in a variety of automotive-inspection problems. Some of the unique methods that have been developed are presented to demonstrate the versatility of nuclear techniques when unusual inspection problems arise: measurement of oil consumption of production engines, monitoring of composition of continuously cast bearing material, detection of internal voids in plastic-foam parts, and measurement of the moisture content of foundry sand.*

The increased emphasis on automotive safety and reliability has in turn increased the demand for new and improved inspection methods for product assurance. While the requirements for the initial materials can be set forth in specifications to be met by the supplier, the quality of the fabricated parts can only be evaluated in the production plant, where certain limitations are placed on the inspection methods that can be employed. For example, inspection methods must be compatible with the process line—in terms of both speed and available space. In addition, simplicity and reliability are also required. In certain instances nuclear techniques provide solutions that are unobtainable or extremely difficult by other methods.

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Specific applications demonstrate the unique capabilities and versatility of nuclear techniques when unusual inspection problems arise—e.g., radiometric oil-consumption measurements of engines, detection of internal voids in plastic-foam parts, and a nuclear sand-moisture gage.

OIL-CONSUMPTION MEASUREMENTS OF PRODUCTION ENGINES

Warranty costs could be reduced if the cause of excessive oil consumption in some automotive engines could be detected on the assembly line. Historically, one of two methods is normally used. The most widely used is drain-and-weigh, in which the crankcase oil is weighed before and after the engine is operated for an extended test period (about 12 hr). In the second method, oil-consumption measurement is based on the use of a calibrated dipstick. Neither of these methods is sufficiently fast or accurate for in-plant measurements.

A radiometric method was developed that requires only a few minutes of engine running time to obtain an accurate measurement. In this method, oil containing a small amount of a radioactive bromine compound is added to the engine crankcase fill.[1] If any crankcase oil enters the combustion chamber during engine operation, the radioactive tracer is converted to HBr and carried out with the exhaust gases, which are passed through canisters where the bromide is captured on porous pellets saturated with an NaOH solution. The amount of radioactivity collected in the canisters is proportional to the rate of engine-oil consumption.

To evaluate the test method, five new 340-in.^{3} engines were examined. One engine was built to standard production specifications. The others were assembled with known defects most likely to occur in production. The engine defects and their resulting oil consumption at 2000 rpm and 20-in.-Hg manifold vacuum are as follows:

1. Standard production engine (no known defects): 1940 ± 200 miles/qt.
2. Engine with top compression ring in cylinder No. 2 installed upside down: 1450 ± 85 miles/qt.
3. Engine with Teflon inserts pulled out of the neoprene valve stem seals and left free on the valve stem (two valves per bank): 1640 ± 70 miles/qt.
4. Engine with a $\frac{1}{16}$-in. slot cut in the lower side of the intake manifold gasket (one defective gasket per bank): 830 ± 87 miles/qt.
5. Engine with a broken expander ring installed behind the oil ring (one defective oil ring expander per bank): 1270 ± 70 miles/qt.

On V-8 engines, two canisters are used—one for each cylinder bank. Figure 1 shows the installation at the Buick Motor Division; the insert shows the structure of the canister and its location in the exhaust system. The total quantity of radioactivity required in the engine oil to achieve the desired sensitivity is 15 μCi which is a general-license* concentration pursuant to Part 30 of the AEC Rules and Regulations.

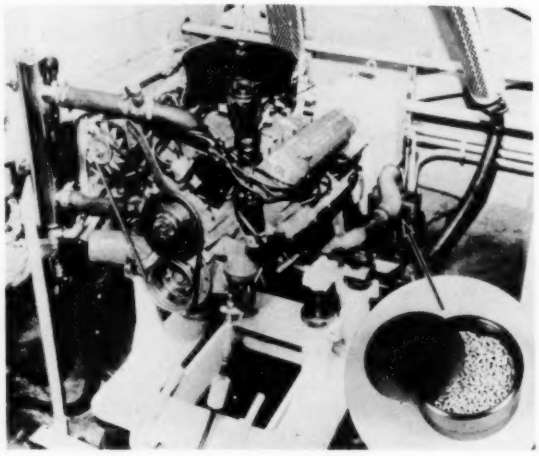

Fig. 1 Arrangement for measuring oil consumption of production engines. Insert shows the structure of the canister and its location in the exhaust system.

The distribution of oil consumption of twenty-five 430-in.^{3} production engines is shown in Fig. 2. Engine No. 1 (687 miles/qt) was installed in a chassis and road tested for 1000 miles; the oil consumption of the road test was 562 miles/qt. When engine No. 6 (738 miles/qt) was torn down and inspected, the number two ring on cylinders 6 and 8 was found to be installed upside down. The engine was re-run after correct reassembly and gave 1468 miles/qt. Thus, with the oil consumption of the standard-production engine as a reference, various defects can easily be detected.

INSPECTION OF ALUMINUM–BABBITT BEARING MATERIAL

General Motors Corp. presently manufactures at its Delco Moraine Division an aluminum–babbitt

*General-license or "exempt" concentrations require no AEC or state license for use.

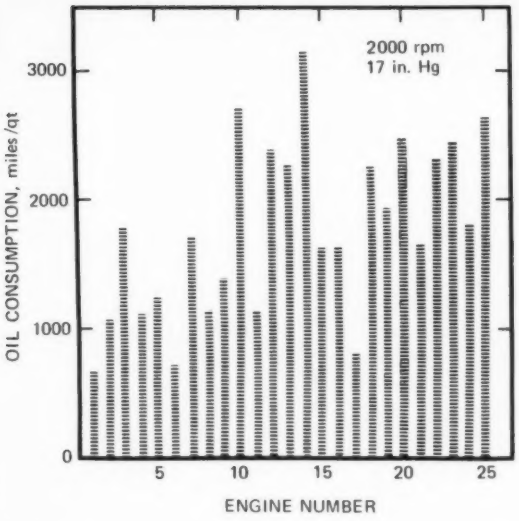

Fig. 2 Distribution of oil consumption of twenty-five 430-in.^{3} engines as determined by the radiometric method.

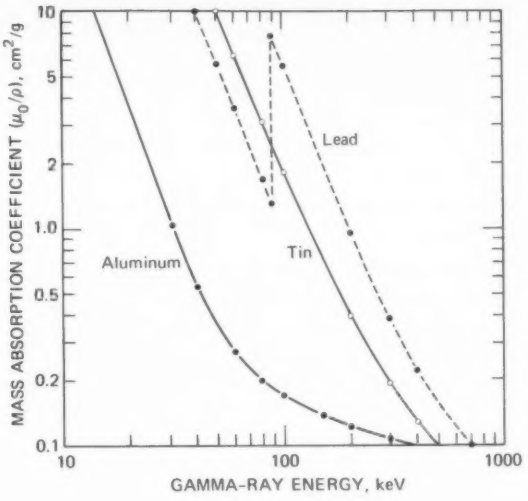

Fig. 3 Mass absorption coefficients of lead, tin, and aluminum.

automotive-sleeve bearing material designated M-390. This material is regarded as a novel concept in bearing technology—the dispersion of an immiscible babbitt phase increasing in concentration through the thickness of a continuously cast aluminum-alloy strip.[2]

In the production of this material, monitoring the total babbitt content of the aluminum alloy as it is cast is important. This babbitt content can be determined by gamma-ray attenuation if proper gamma-ray energies are selected.

The mass absorption coefficients (μ/ρ) for lead, tin, and aluminum as a function of gamma energy[3] are plotted in Fig. 3. Upon the basis of this information, a 350-mCi ^{241}Am source was selected, which has a favorable half-life of 458 years and emits gamma rays of 60 keV. At this energy level, the gamma radiation is readily absorbed by the heavy elements but is not significantly attenuated by the aluminum. The count rate will, therefore, indicate the total babbitt content in the aluminum strip.

The production-line system for continuous monitoring of the total babbitt content in the aluminum strip as it is cast consists of an isotope exposure unit, a G–M detector, a count-rate meter, and a millivolt strip-chart recorder—arranged on the production line as shown in Fig. 4. The unit is designed to monitor babbitt content in the range of 5 to 7% with a sensitivity of ±0.05%.

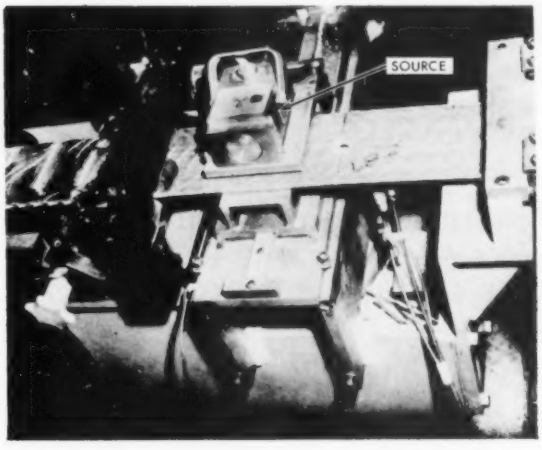

Fig. 4 Source and detector arrangement on M-390 production line.

CARBURETOR-FLOAT INSPECTION

Another application which demonstrates the versatility of nuclear techniques is a method for production-line inspection of low-density plastic-foam carburetor floats. Occasionally subtle changes in the manufacturing conditions produce floats with internal voids and weakened cell walls.

When looking for voids in many materials, radiography is automatically considered. In this case, how-

ever, large production volume and low sensitivity eliminate radiography as an inspection method. Neither can gravimetric determinations be used because the normal variations in the weights of the floats are greater than the changes expected in defective floats. Although a radiation-transmission approach might at first be rejected for these same reasons, it could be used if an enhancement in sensitivity could somehow be effected.

The approach finally chosen was to increase the relative radiation attenuation of defective floats by use of a radiopaque fluid. A 50% saturated solution of barium bromide or zinc chloride could be used. The radiation source was ^{241}Am, and the detector was a G–M tube. Batches of floats were vacuum-impregnated with the radiopaque fluid and then passed between the radiation source and detector to provide a signal that would cause those floats which had absorbed a significant quantity of fluid to be rejected.

A reproduction of the strip-chart recording of one series of laboratory tests (Fig. 5) demonstrates the signal level obtained from defective floats. The radiation attenuation record is compared with radiographs of the inspected floats to show the distribution of fluid in the defective floats. Examination time was approximately 1 sec per float, a rate that is compatible with the production volume.

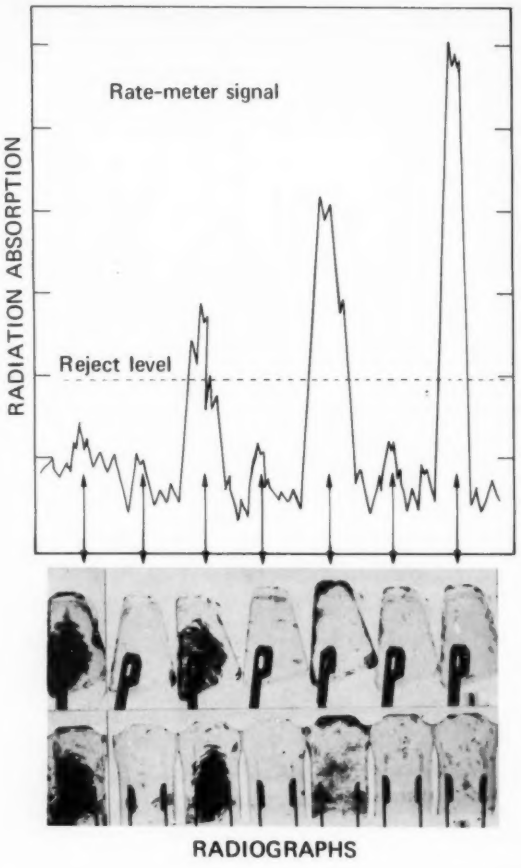

Fig. 5 Strip-chart recording (top) of a group of floats and radiographs (bottom) to show sensitivity levels.

GREASE CONTENT IN FRONT-WHEEL ASSEMBLIES

Front-wheel disk-brake assemblies are produced on an automated line. The assembly includes the steering knuckle and spindle assembly, splash shield, hub and disk, and two bearings with their grease supply. A total of approximately 30 g of grease is supplied to the two bearings through an automatic metering system. Any malfunction of the grease system can result in inadequately greased bearings—a condition that is not easily detected by visual inspection. Therefore nonvisual inspection methods that could be automated had to be considered.

The presence or absence of bearing grease could be easily determined by adding appropriate quantities of radioactive material to the grease; however, such an inspection procedure in a production plant would be limited by two important factors: First, maximum sensitivity cannot be achieved because the fact that the parts are quickly released from the plant allows only the use of exempt concentrations of radioactive materials under AEC regulations. Second, only small percentages of foreign materials can be added to the grease without altering chemical and physical characteristics. Within these limitations, a ^{75}Se diphenyl tag offered the best combination of sensitivity, radiation safety, and physical compatibility with the grease.

Two inspection stations were needed on the assembly line—one station following each bearing installation. At the first inspection station one of two conditions would exist (Fig. 6): Case 1 would be for a dry inner bearing and Case 2 for a properly lubricated inner bearing. After the inner-bearing inspection, the steering knuckle, spindle assembly, and outer bearing are installed. At the second inspection station, Case 3 is for a dry outer bearing and Case 4 is for a properly lubricated outer bearing.

Figure 7 shows the levels of activity detected for the four possible inspection cases illustrated in Fig. 6.

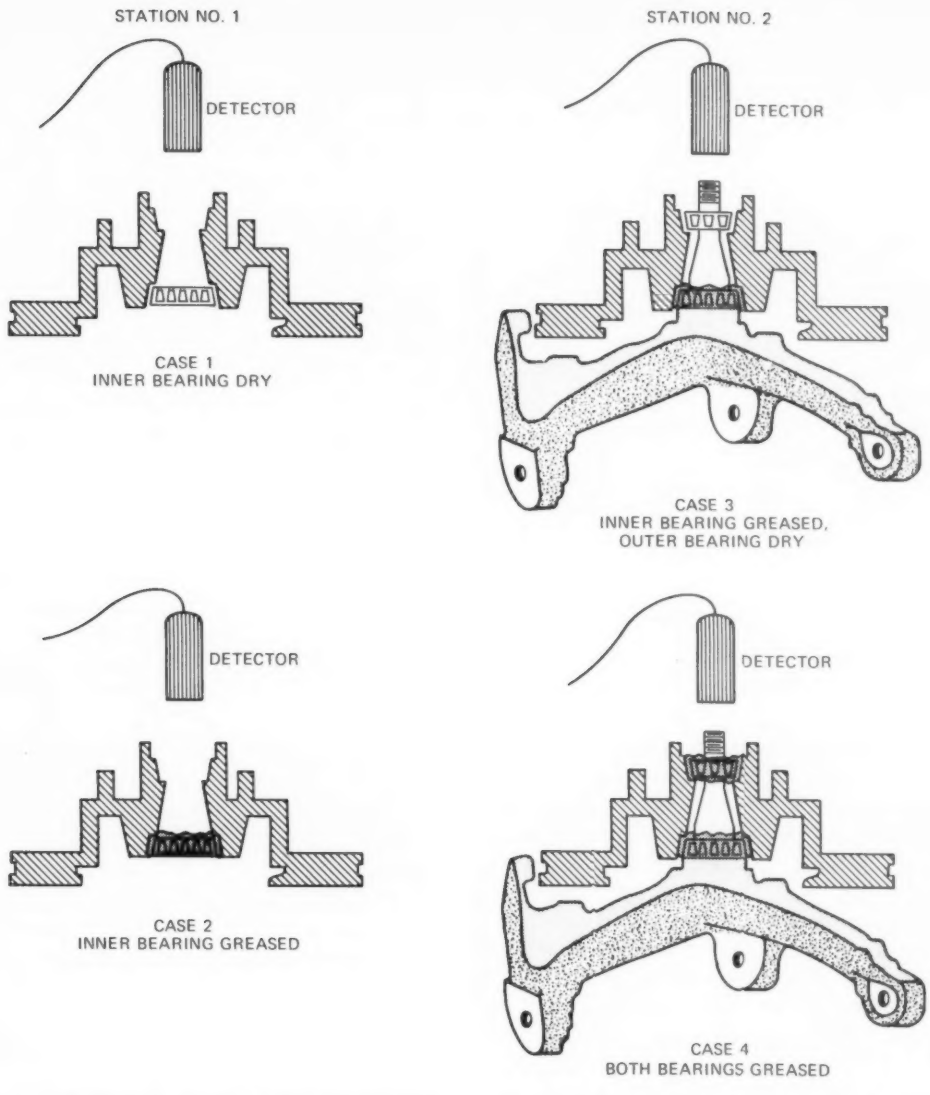

Fig. 6 Illustration of the four possible conditions that might occur at two inspection stations. The wheel and disk assembly is shown in cross section to reveal the roller bearings and their condition.

Note how the spindle shields the radiation from the inner bearing at the second inspection station so that only the outer bearing is inspected.

NUCLEAR SAND-MOISTURE GAGE

Foundry sand comprises sand mixed with small amounts of clay, sea coal (powdered coal), and water. The water content is critical because a precise amount is required for proper conditioning of the sand, and small deviations can result in inferior molds and castings. A typical moisture content would be 2.8 ± 0.05 wt.% water.

The foundry sand is made up of dry sand (reclaimed from used molds) and clay, sea coal, and water (spent in the casting process), which are added in the form of a slurry and mixed with the sand by a muller. In the past, the amount of water added to the sand was determined by an operator who "felt" the sand coming from the muller and judged its moisture content. As a

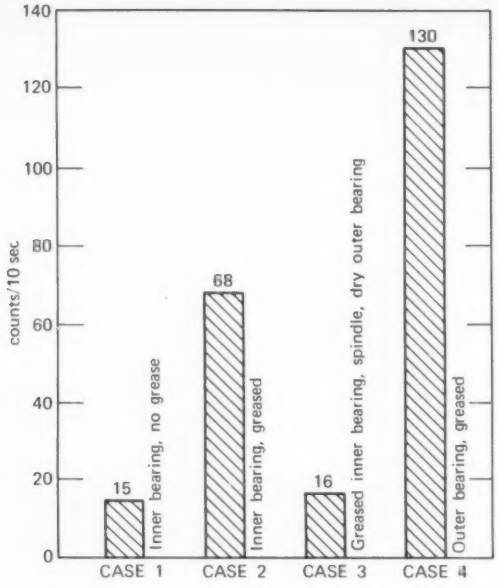

Fig. 7 Radiation levels under the four conditions presented in Fig. 6.

rule, good results were obtained by this procedure, but unacceptable departures from the required moisture content often resulted because of unskilled operators or sudden variations in spent-sand quality entering the muller.

As fast neutrons pass through materials, they lose energy by collision with the atomic nuclei so that eventually they become thermal neutrons—neutrons of very low energy. Since hydrogen nuclei moderate the neutrons much more effectively than other nuclei, the thermal-neutron flux in a material is usually proportional to its water content.

However, for foundry sand with moisture content in the range of 2 to 4 wt.%, the thermal-neutron flux is not directly related to the sand-moisture content. In the range of 2 to 3.5 wt.% moisture, the thermal-neutron count rate actually decreases as the moisture of a fixed volume of sand increases, whereas above 4 wt.% moisture, the neutron count rate becomes substantially a linear function of moisture content. This decrease in neutron count rate as the moisture is increased in the range of 2 to 3.5 wt.% is due to a decrease in sand density that, in turn, is attributed to the clay. When clay absorbs water, it expands considerably more than can be accounted for by the volume of water added. Since the thermal-neutron detector effectively senses the hydrogen content in a fixed volume of sand, the neutron count rate will decrease as the density decreases.

The thermal-neutron count rate can be adjusted mathematically by taking into account the sand density to provide a normalized thermal-neutron count rate that is quite linear—continuously increasing as the moisture content increases and representing the thermal flux as it would be if the sand density remained constant.

Figure 8 depicts the system developed to measure the water content and the density of foundry sand continuously flowing through a chute. A probe within the chute carries a fast-neutron source and a slow-neutron detector to measure the amount of water. A gamma source in the probe and gamma detectors outside the chute measure the sand density. The neutron and gamma detector signals are arithmetically combined to give a signal proportional to percent moisture by weight. The signal is then used to control water additions to the sand.

Figure 9 illustrates the advantage of the nuclear sand-moisture gage. During Shift A, typical deviations in sand-moisture content occurred prior to installation of the gage. During Shift B, the quality of the sand was much more uniform with the gage in operation.

OTHER INSPECTION TECHNIQUES

With the development of new radioisotope sources, nuclear techniques that once were restricted to laboratory use are becoming available for in-plant purposes. Two specific examples are the well-known neutron activation analysis and the more recent X-ray fluorescence analysis with radioisotope sources.

Neutron Activation Analysis

Just how does neutron activation analysis compare in effectiveness with other types of analysis, such as spectroscopic, gravimetric, and colorimetric? The bar graph in Fig. 10 shows that activation analysis is potentially more sensitive than the other methods by an order of magnitude. It shows further that the sensitivity can be enhanced by increasing the neutron flux.

Some of the advantages of activation analysis are its speed and the fact that it does not cause damage to the specimen. The major drawback to date has been the need for nuclear reactors to supply the high neutron-flux levels. Isotopic neutron sources have always been attractive because of their simplicity and maintenance-free operation. The characteristics of a

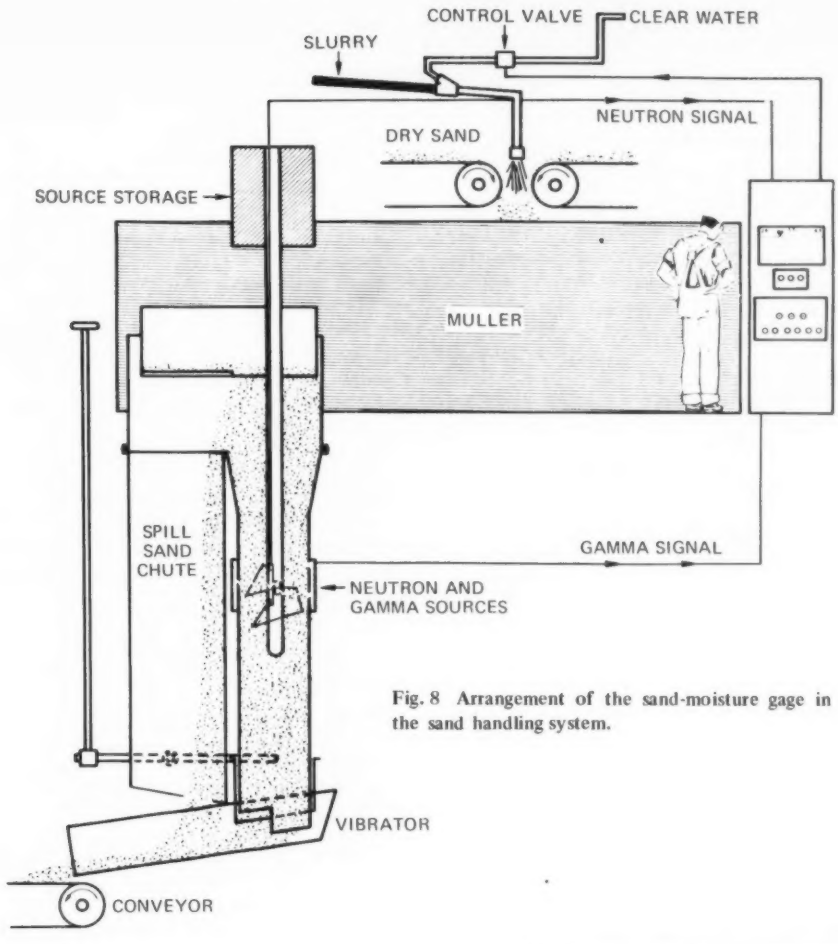

Fig. 8 Arrangement of the sand-moisture gage in the sand handling system.

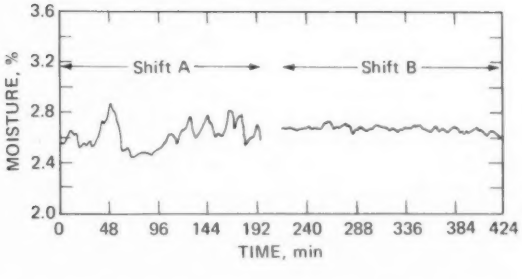

Fig. 9 Control of sand-moisture content during Shift A by operator only. During Shift B, operator had the aid of the sand-moisture gage.

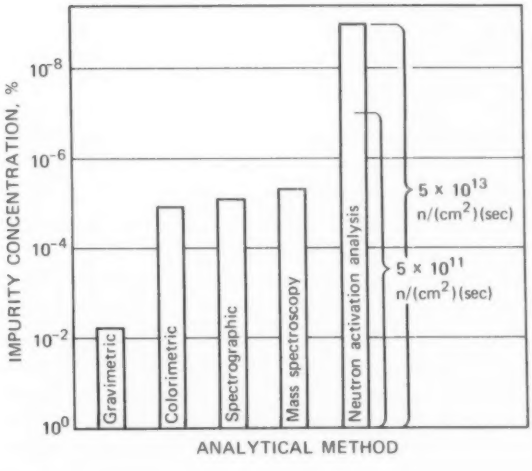

Fig. 10 Sensitivity comparison of neutron activation analysis with other types of analysis.

few neutron sources are listed in Table 1. However, the relatively low flux, large volume, and/or short half-life of these sources have severely limited their use.

With the development of ^{252}Cf neutron sources,[4] in-plant activation-analysis systems are becoming a possibility. The nuclear properties of this isotope are ideal with a reasonable half-life of 2.6 years and an output of 2.3×10^{12} n/(sec)(g). The Atomic Energy Commission is presently encouraging the development of commercial applications of ^{252}Cf.

Table 1 Characteristics of Neutron Sources

Source	Half-life, years	Output, n/(sec)(g)
$^{239}Pu(Be)$	24,360	1.3×10^5
$^{241}Am(Be)$	458	6.9×10^6
$^{238}Pu(Be)$	89	4.7×10^7
$^{244}Cm(Be)$	17.6	2.1×10^8
^{252}Cf	2.6	2.3×10^{12}

X-Ray Fluorescence Analysis with Radioisotope Sources

In fluorescence analysis, gamma or X-ray radiation is used to excite the specimen. This radiation interacts with the electrons of the target atoms—displacing some of the electrons from their orbits. When other electrons fill these vacancies, X rays of discrete energies appear which are characteristic of the target atoms. The sensitivity of the technique depends largely upon the source of the incident radiation.

There has been a great deal of attention directed to this technique lately because of the development of new radioisotope excitation sources. Up until this time, the lack of proper source energies and intensities has greatly limited its potential applications. Now a wealth of literature is appearing on the development of X-ray fluorescence both as a laboratory method and as a nondestructive testing procedure.[5-8] The radioisotope sources are virtually maintenance free; they are relatively inexpensive, and, because of their small size and the absence of auxiliary power requirements, they are suited to in-plant applications. Some of the radioisotope sources which emit low-energy electromagnetic radiation suitable for X-ray fluorescence analysis are listed in Table 2.

Table 2 Low-Energy Gamma Emitters Suitable for Fluorescence Analysis

Nuclide	Half-life	Radiation energy, keV
^{109}Cd	1.3 years	22
^{145}Sm	1.0 year	39
^{55}Fe	2.7 years	5.9
^{125}I	57 days	27
^{241}Am	458 years	60(40%) 10 to 30(35%)

Since the low-energy radiation penetrates only to a shallow depth and the intensity of characteristic X rays generated in a material is proportional to the number of target atoms present, these sources are ideally suited for surface composition and coating-thickness measurements. (FEM; PBS)

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Activation Analysis Program at Gulf Radiation Technology: The First Decade

By H. R. Lukens*

For a decade, the Activation Analysis Group at Gulf Radiation Technology, formed in 1961, has offered commercial and contract services related to the applications of activation analysis. Since an earlier description,[1] the facilities have been supplemented with several useful items, including a 36-cm Ge(Li) 4096-channel gamma-ray spectrometer, additional pneumatic sample-transfer systems, and BF_3 and Čerenkov counters. In commercial analytical work as well as in research and development, the Group has encountered numerous interesting topics, which are reflected in its approximately 200 publications. This article discusses some of the more interesting ones.

RESEARCH AND DEVELOPMENT

Ge(Li) Detectors

The Ge(Li) detector has had a great impact in extending the capability of purely instrumental activation analysis for multielement determinations. For example, we can now determine all the rare earths in single radiochemically separated rare earth fractions of a sample (soils, ores, etc.) by Ge(Li) spectrometry. The multielement-scan procedure for simultaneous estimation of major, minor, and trace elements has proved to be very effective when the Ge(Li) detector is used, and full advantage of this has been taken in our research on the uses of neutron activation analysis for identification of materials. It is not uncommon to determine more than 20 elements in a single sample (one soil sample yielded 29 elements).

This does not mean, however, that radiochemical separations have been abandoned. For example, mercury is determined in fish by a 30-min irradiation with a thermal-neutron flux of 2×10^{12} n/(cm^2)(sec), followed by a quick radiochemical separation and a 10-min count with a NaI(Tl) detector. This is not only more sensitive than the Ge(Li) technique but is much faster, since a decay time of several days must be allowed before the 1-hr count is made with the Ge(Li) detector–spectrometer.

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Criminalistics

An important area of application of the activation analysis–Ge(Li) detector technique is in criminalistics. Comparison of paint, hair, or other material taken from the scene of a crime with that from a suspect is often the basis of police action. At one time, attempts were made to interpret a simple color comparison of paint samples; however, it is now possible to obtain an average of 15 points of comparison by neutron activation analysis of a paint. Our investigation of the trace-element pattern ("fingerprints") of a stratified sampling of the population of painted objects has provided the basis for interpretation of such comparisons with an extremely high degree of certainty. It had been anticipated that trace elements in many of the materials investigated would display normal or log-normal concentration distribution functions, and it would then be possible to formulate probabilistic statements by the multivariate normal (MVN) distribution model. This has not proved to be true for paint, paper, or bullet lead, but, for paint and paper, the population sample data could be used to define empirical distribution functions and generate the desired statements with a multinomial model.

The neutron activation analysis method for measuring gunshot residues in paraffin handlifts was developed at this laboratory in the early 1960s. However, not until 1970 were sufficient data gathered on blanks and on hands of persons who had fired common calibers of handguns to permit formulation of necessary statistical statements. It was found that the bivariate normal distribution (BNV) model could be used to treat the two variates of the method, barium and antimony. An apparent violation of the BVN model was presented by the finding that neither barium nor antimony values followed Gaussian statistics. This was resolved by recognition of differences in thermodynamic properties of compounds of the two elements, which became important because of temperature and pressure differences between handguns. With due allowance for the basic difference in weights of the two elements in unfired cartridges, it was verified that the sum of the two elements is distributed normally in handlifts.

Oil-Spill Identification

In the use of activation analysis for identification of an oil-spill source, trace-element patterns were obtained for several hundred samples of oil, mostly from marine oil spills. After determination of the elements whose concentrations are stable under environmental conditions, the trace-element patterns were subjected to statistical analysis. A method was obtained by which the degrees of certainty as to whether two given samples (slick and suspected source oil) are matched or unmatched could be determined. It was found that in 99.5% of cases the obvious interpretation has a vanishingly small chance of error. In the treatment of oil data, a number code system was devised to describe the trace-element patterns, and an objective method of comparing pattern number-code pairs—a simple computation of numerical differences—was tested. The difference values for matched pairs of oils follow a binomial distribution, while those of unmatched pairs follow an empirical distribution curve.

Biological-Tissue Analysis

For determining the calcium content of monkey bones in vivo, we prepared accurate bone models as standards and irradiated them and unknown samples over a thermal column of the TRIGA Mark I reactor at a flux of 10^7 n/(cm^2)(sec) for 10 min (approximately 1 rem) and counted for 10 min with a 5- by 5-in. NaI(Tl) counter coupled to a 400-channel pulse-height analyzer. The low-flux measurements agreed well with values obtained by dissolving the bone samples and determining the calcium by both atomic absorption and standard high-flux neutron activation analysis:

	Ca, g		
Sample	In vivo NAA	Standard NAA	Atomic absorption
Vertebra A	1.47 ± 0.08	1.47 ± 0.05	1.54 ± 0.05
Vertebra B	1.52 ± 0.10	1.67 ± 0.02	1.58 ± 0.05
Femur, average	8.21 ± 0.31	8.38 ± 0.10	8.20 ± 0.20

Analyses of tissues have been of considerable interest in both routine and research work. While some requirements have involved the determination of only one element—e.g., mercury—many have involved the determination of a number of elements. A clear definition of the sensitivities, precisions, and accuracies required for the various elements must be obtained in order to design an appropriate procedure. In an early case where we had been given verbal assurance that ±100% relative precision would be satisfactory, we subsequently received written insistence on ±10% relative precision.

The elements Ca, Co, Cd, Mn, Zn, V, and Se were determined in 100-mg samples of arterial tissue to satisfy the requirements of a customer. Samples were irradiated 30 min at a flux of 1.8×10^{12} n/(cm^2)(sec) and then digested in hot sulfuric acid containing all carriers but calcium. The digested sample was diluted, the calcium sulfate centrifuged off, and the solution made ammoniacal. After separation of the elements into groups by liquid–liquid extraction, gamma-ray spectrometry was carried out on each group, using the usual indicator isotopes except where the more sensitive 10-min ^{60m}Co and 57-min ^{81m}Se were used instead of 5-year ^{60}Co and 120-day ^{75}Se, respectively. It was possible to process—irradiation, chemistry, and counting—four samples in less than 2 hr by using four spectrometers for the counting. The element most severely penalized was vanadium, which was measured as 3.8-min ^{52}V; nevertheless, a sensitivity of 40 parts per billion of V was achieved, which was more than adequate for the purpose of the work.

Very-Short-Lived Isotopes

Rapid, pneumatic sample-transfer systems were installed for the observation of very-short-lived isotopes. With a 0.5-sec delivery time from reactor to counting station, it was feasible to observe 0.8-sec ^{207m}Pb. Where a 15-msec 10^9-W reactor pulse was used to activate a sample and a 3- by 3-in. NaI(Tl) detector was used to count the sample, there were 262 counts/sec per milligram of lead. Installation of a Lucite Čerenkov counter in the fast-transfer system allowed the observation of Li, Be, B, and other elements that give rise to short-lived hard-beta emitters. The observation of 0.85-sec ^{8}Li subsequent to a pulse was particularly impressive—the sensitivity for Li was 80 pg with the ^{7}Li$(n,\gamma)^8$Li reaction. Despite the impressive performance of the system, there has been little demand for Li determinations. However, the ^{11}B$(n,\alpha)^8$Li reaction has been useful for the measurement of boron in several materials.

Bremsstrahlung Activation

While 0.8-sec ^{6}He from the ^{9}Be$(n,\alpha)^6$He reaction provides a practical means of determining beryllium,

we can use our accelerator-produced bremsstrahlung to determine the element by the well-known photodisintegration reaction. The latter route is specific for beryllium, provided a bremsstrahlung of <2.22 MeV is used. At that energy, deuterium photodisintegrates. Also, if we use a counter for the samples irradiated at these energies rather than measuring the neutrons, (γ,γ') products can be observed. Low-energy (<6 MeV) bremsstrahlung irradiations have proved useful in the determination of Br, Ag, Er, Hf, Ir, and Au.

At higher bremsstrahlung energies, useful photonuclear reactions are produced, which have been of particular value in the determination of C, O, F, and Ni. Perhaps the determination of fluorine has been the most useful of all our photonuclear applications, since practical sensitivities of 0.1 ppM can be achieved in many—particularly organic—materials. Fewer interferences are generated from the matrix than in the inherently more sensitive reactor-activation method for fluorine. Typical applications have been the determination of fluorine in teeth, petroleum, and crops.

Delayed-Neutron Activation Analysis

The chief advantage of the purely instrumental delayed-neutron method is its speed, but the minimization of material loss due to processing of samples without extensive chemical treatment is also important. The method has been useful in filling the increased demands for the determination of fissile and fertile materials which have accompanied the increase in nuclear power. We equipped our fast-neutron system with a BF_3 counter several years ago for use in determining delayed-neutron emitters produced in fission with a 30-sec irradiation in the core of the TRIGA Mark I reactor, followed by counting at 1 to 40 sec after the irradiation. A sensitivity of <1 μg for natural uranium and of 50 μg for thorium was achieved.

The system has been used successfully to determine uranium in ores. When three reference ores supplied to us as unknowns by the AEC's New Brunswick Laboratory were analyzed by this method, the results compared well with those of round-robin analyses of the same ores by wet chemistry in more than 15 laboratories:

	U_3O_8, wt.%	
Sample	Wet chemistry	Instrumental
119	0.175 ± 0.003	0.174 ± 0.004
131	0.384 ± 0.004	0.382 ± 0.007
133	0.290 ± 0.003	0.291 ± 0.006

An interesting application now much used, with an improved system having a six-tube BF_3 counter, is the determination of uranium and thorium in reactor fuel and fertile material, where extreme accuracy and precision are required. In large (1- to 10-g) samples of enriched nuclear fuel containing as much as 300 mg of ^{235}U, we must account precisely for the factors that perturb the neutron flux in the sample. This is done by measuring a series of sample aliquots of increasing weight and plotting the log count per unit weight vs. the aliquot weight, doing the same to carefully prepared standards. The result is obtained from the sample/standard ratio of zero-weight intercepts. Despite the inevitable small discrepancies between the standard and sample matrixes, we have found the zero-weight intercepts of these and a pure U_3O_8 standard differing by only 0.3% (relative). In another program, small amounts of fissile material in large waste drums were determined satisfactorily by the delayed-neutron technique (with suitable modifications).

Computerization

As might be expected, the use of computers to handle data-reduction tasks has become routine. In addition to an array of codes to handle the various tasks for reducing gamma-ray spectrums, least-squares codes (e.g., to fit the specific activity vs. sample weight curves discussed above) and various special programs have been written. For example, trace-element patterns in all pairs of oils in a sample set are compared by computer.

SERVICE WORK

Day-to-day problems associated with carrying out commercial analyses on widely different materials for a large variety of elements usually include selecting an economical, but satisfactory, analytical protocol for a task that may not recur. Often this must be done over the telephone to satisfy the customer's need for an immediate price quotation. It is not uncommon to find that the customer's sample description did not include an interfering element, and we usually consider the contingency that some of the more common interferences will be present, based on our experience plus rather complete tables of the elemental compositions of plants, alloys, rocks, etc.

Many customers are cognizant of the sampling problem and often supply representative and weighed samples, sealed in vials and ready for irradiation. Only

rarely is an appropriate sampling protocol not developed. For example, in the case of the arterial tissue mentioned above, agreement was reached that we would trim samples with quartz, rather than metal, knives in order to decrease the possibility of contaminating the sample. However, there have been instances where polyethylene bottles—originally filled with light hydrocarbons—were received empty; unmarked samples have been received; and in some cases rush samples have been shipped by ground transportation, only to arrive many days after the results were required. In most instances, however, information concerning the nature of the problem behind the need for analysis is forthcoming, often accompanied by a request for advice or confirmation regarding the analytical specifications. (MG)

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An Evaluation of ^{252}Cf as a Neutron Radiography Source*

By J. P. Barton and M. F. Klozar †

Supplementary Keywords: *source, neutron; design.*

Abstract: *Experiments have been performed to test the suitability of the spontaneous-fission neutron source ^{252}Cf for neutron radiography. With a water moderator, the measured ratio of fast-neutron yield-to-peak thermal-neutron flux is at least a factor of 2 better than obtained with other sources. Divergent cadmium-lined collimators have been studied over a range of ratios from 1 : 7.5 to 1 : 30. The effects of collimator ratio on (1) exposure rate, (2) contrast of selected materials, and (3) resolution have been evaluated. As the collimator ratio is increased, the exposure rate decreases at a rate less than that predicted by the inverse-square law, and, although contrast decreases, the overall detail in the neutron radiograph increases rapidly. The effects of changes in beam-extraction geometry and in the collimator dimensions for a constant collimator ratio are shown. In particular, the introduction of a 5-cm air gap between source and collimator in tangential geometry is more advantageous to radiographic resolution than a factor of two in collimator ratio. Detection methods demonstrated include LiF–ZnS scintillator and gadolinium-metal conversion screens with neutron exposures of 7×10^5 n/cm² and 1.3×10^8 n/cm², respectively.*

Over 50 nuclear reactors have been equipped for neutron radiography. Now the first application facilities are being established on the basis of nonreactor sources such as small accelerators and radioactive isotopes. Because of the low intensity of neutrons from convenient nonreactor sources, the quality of neutron radiography will not be as good as that obtained by reactor beams.[1] However, neutron radiography can provide information unobtainable from X rays, and a number of applications can be foreseen where low-quality neutron radiography would be sufficient (e.g., blocked oil or water pipes, plastic insulation, and gasket positioning).

One isotopic neutron source of interest is ^{252}Cf (Refs. 2–4). This material, generated by successive neutron buildup in a nuclear reactor, yields neutrons through spontaneous fission. The characteristics of the source are given in Table 1. The U. S. Atomic Energy

Table 1 Characteristics of ^{252}Cf

Neutron yield	2.34×10^{12} n/(sec)(g) 4.4×10^9 n/(sec)(Ci)
Neutrons per fission	3.76 (average)
Neutron energy	2.3 MeV (average)
Alpha decay half-life	2.73 years
Spontaneous fission half-life	85.5 years
Effective half-life	2.65 years
Gamma dose rate	1.3×10^{13} photons/(sec)(g)
Heat generation	38.5 W/g

*Contains material from a paper presented at the Winter Meeting of the American Nuclear Society, San Francisco, Nov. 30–Dec. 4, 1969, and from extracts first published in *Californium-252 Progress*, No. 1, pp. 20-21, October 1969, and No. 2, pp. 20-25, January 1970.

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Commission (AEC) has predicted that, if the material were manufactured in an optimized program, sources could be made commercially available in 10 years at a cost of about $1/μg.

This article presents the results of initial trials to determine whether or not the source is suitable for radiographic applications. The study considers thermal-neutron radiography (as opposed to fast-neutron radiography). The source was manufactured at the AEC Savannah River Laboratory and was loaned to Argonne National Laboratory (ANL) for radiography trials as part of a comprehensive program to determine the market potential in applications such as medical neutron therapy, mineral exploration, and nuclear safeguards.

SOURCE AND EQUIPMENT

The ^{252}Cf source had a measured neutron yield of 3.2×10^8 n/sec ± 2% in March 1969. Assuming a relationship of 2.3×10^6 n/(sec)(μg), the equivalent ^{252}Cf content would be 140 μg. The radioactive material was contained in a platinum pellet (~1.8 mm in diameter and 2 mm in length) and was doubly encapsulated in stainless steel, with external capsule dimensions of 9.4 mm diameter and 13.5 mm length. The containment capsule had a protruding screw thread with an attached rod for manipulation. For convenience, the experiments were performed in a kilocurie hot cell with shielding adequate for neutron sources of 10^9 n/sec. During experimental measurements, the source was centered in a small water tank (Fig. 1); when not in use, the source was stored in a shield of boronated paraffin of ~30 cm radius. The radiation dose rates at the surface of this shield were 400 mR/hr neutrons and 100 mR/hr gamma.

NEUTRON MODERATION

The requirement of the neutron moderator is to provide the highest possible peak thermal flux at one small region (the divergent collimator input aperture).[5] A slightly lower thermal flux available over a much larger volume would not, in general, be of value.

Hydrogenous moderators are effective because they moderate the neutrons rapidly with the fewest collisions in a concentrated region near the central radiating source. For the particular case of ^{252}Cf, Cutforth[6] has argued that hydrogenous moderators, such as H_2O or $[CH_2]$, are to be preferred to others, including beryllium. Cason uses small pieces of ZrH_2 around a ^{252}Cf source as moderator.[4] For convenience in these studies, ordinary water was used as moderator although high-density polyethylene should be even more effective (the hydrogen density of water is 6.7×10^{22} H atoms/cm^3; the highest theoretical hydrogen density of polyethylene is 8.34×10^{22} H atoms/cm^3). The dimensions of the moderator system (559 mm diameter and 457 mm high) were chosen so that, as far as the peak central flux was concerned, the dimensions were effectively infinite (i.e., the addition of further moderator material beyond these dimensions could increase the peak central flux by only 10%).[6]

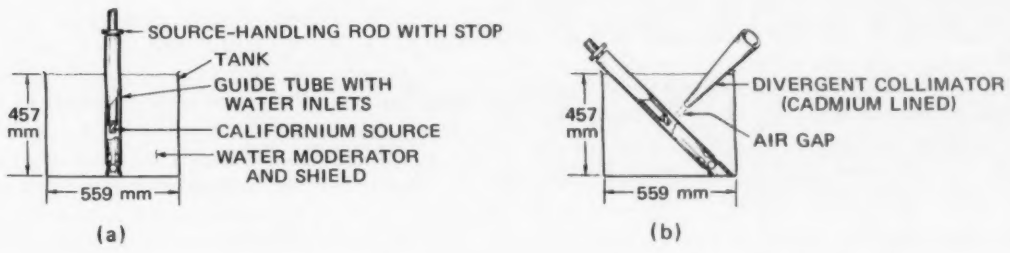

Fig. 1 Experimental arrangements for ^{252}Cf source thermal-neutron flux plots (*a*) and radiography trials (*b*).

The magnitude and distribution of the thermal-neutron flux were measured with the ^{252}Cf source in the arrangement shown in Fig. 1(*a*). Indium-foil activation techniques were employed, using bare and cadmium-wrapped foils in separate irradiations. The indium was 0.25 mm thick, the cadmium 0.5 mm thick, and the activities were measured in a calibrated 2π gas-flow beta-ray counter. The results are shown in Table 2.

The peak thermal-neutron flux obtainable from a given yield of fast neutrons from ^{252}Cf is high. The

Table 2 Thermal-Neutron-Flux Distribution for ^{252}Cf in Water

Distance from source, mm	Thermal-neutron flux, 10^6 n/(cm^2)(sec)	Ratio of fast-neutron yield/thermal-neutron flux, cm^2	Cadmium ratio: total response of 0.25-mm-thick indium foil/cadmium-covered response
25	4.44	76	10.8
38	3.88	87	12.1
51	3.15	107	12.8
63	2.53	133	14.5
76	1.98	170	15.6
89	1.48	227	17.2

measured ratio of fast-neutron yield [n/(sec)(4π)] to thermal-neutron flux [n/(cm^2)(sec)] was about 100 for ^{252}Cf in water, whereas previous measurements for other sources gave values of 200 for ^{241}Am–^{242}Cm–Be and 1000 for ^{241}Am–Be and 14-MeV neutrons (from the D–T reaction). The results for ^{252}Cf, using indium-foil methods, are in agreement with the results of Magnuson[7] for copper-foil techniques.

Table 2 also shows that the peak thermal flux and the best cadmium ratio occur close to the source. The rate of decrease of thermal flux with increasing radius is rapid. From a radius of 25 mm to one of 100 mm, the flux decreases by more than a factor of 3.

COLLIMATION

The beam extraction and collimation in these experiments were achieved with a series of air-filled conical flight tubes, inserted to "look" at the region of peak thermal flux, as shown in Fig. 1(*b*). The divergent collimator system was chosen because, with small-source techniques, it is particularly important to minimize both moderator displacement and neutron absorption by collimator material near the effective thermal-neutron source.

The aluminum conical tubes were lined with 1-mm-thick cadmium to prevent thermal-neutron leakage inward from the water. The neutron-opaque lining therefore provides definite dimensions to the collimator. Cadmium was used, in preference to boral or to borax and lead, because of the convenience in experiments that involved frequent changes in collimator shape and size. However, once the optimum collimator configuration has been determined, a construction using boral or similar material rather than cadmium might be recommended because of good epithermal-neutron absorption and small gamma-ray contamination.

EXPOSURE ARRANGEMENT

An extensive series of neutron-radiograph exposures has been made to evaluate quantitatively the performance of the system. All exposures (Figs. 2 to 10) are for the same detection system—namely, a LiF–ZnS(Ag) scintillator converter system and Kodak Royal Blue film. A 0.25-mm-lead filter was used in front of the aluminum cassette, and the exposure position was shielded against scattered radiation by 50-mm-thick boronated paraffin backed up by 0.5-mm-cadmium and 0.25-mm-lead foils. Film transmission densities were recorded using a Macbeth–Ansco densitometer with a 4-mm spot size to integrate over a suitable area of the rather grainy film.

COLLIMATOR RATIO

Neutron radiographs were made using three collimators, each of a different ratio (inlet aperture : length). The resulting radiographic film density as a function of exposure time is shown in Fig. 2. A surprising result is that exposure times corresponding to a given radiographic density are not proportional to the square of the collimator ratio, as would be calculated from inverse-square-law considerations. For example, for a film density of 2.0 units, corresponding exposure times increase approximately linearly with

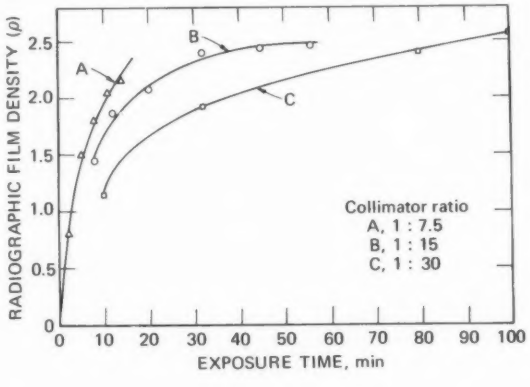

Fig. 2 ^{252}Cf neutron-radiograph transmission density as a function of exposure time for different degrees of collimation. (Collimators of fixed length, 380 mm; collimator input aperture just below source.)

the collimator ratio, rather than with the square of this ratio. The reasons may be twofold: first, the neutron-radiographic density may not be entirely due to collimated thermal neutrons; second, since the collimator input aperture is different for each of the three collimators, the effective source flux will be different in each case. The smaller diameter input (1 : 30 collimator) will not only look more precisely at the peak thermal flux but will also cause less flux depression in the source region than the large-input-diameter collimator.

The overall film density is not as important as the actual quantity of information in the radiograph. One way to obtain a quantitative measurement is to take radiographs of a system image-quality indicator (IQI). This IQI contains different absorbers in a known range of thicknesses as well as high-contrast resolution tests (cadmium wires and holes) and low-contrast resolution tests (plastic wires). Construction of the IQI is described in detail elsewhere.[8] It is most important that all radiographs be exposed to about the same density when performing quantitative tests with the system IQI. The densities for these experiments were confined to the range 1.95 ± 0.05 units.

The appreciable radiographic contrast obtained for thin sections of cadmium can be seen in Fig. 3. This

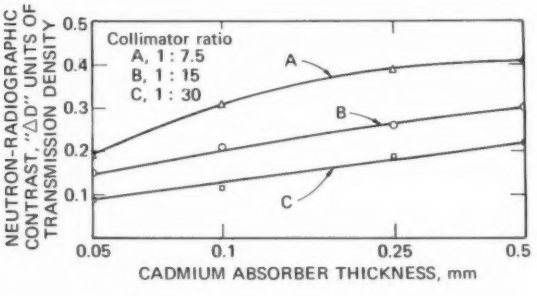

Fig. 3 ^{252}Cf neutron-radiograph contrast as a function of cadmium absorber thickness for different degrees of collimation. (Collimator of fixed length, 380 mm; collimator input aperture immediately below source.)

graph shows the rate of increase of contrast as the cadmium thickness increases for the three collimator ratios. A change in the collimator ratio from 1 : 30 to 1 : 7.5 causes a greater increase in contrast than a 10-fold increase in absorber thickness (from 0.05 to 0.5 mm). In Fig. 4, the neutron-radiographic contrast is compared for four selected materials.

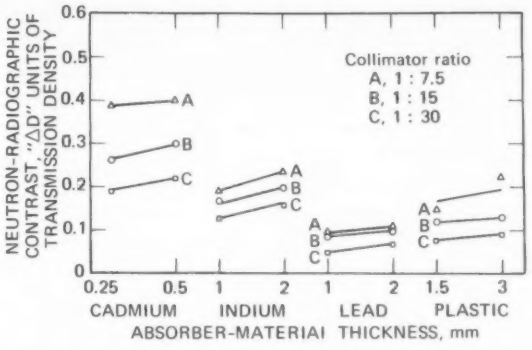

Fig. 4 ^{252}Cf neutron-radiograph contrast as a function of absorber material for four materials and for three degrees of collimation. (Collimators of fixed length, 380 mm; collimator input aperture immediately below source.)

Cadmium is, of course, a strong absorber of thermal neutrons, a medium absorber of gamma radiation, and a weak absorber of epithermal neutrons. Indium has a relatively weaker response to thermal neutrons, a response to gamma radiation similar to cadmium (cadmium and indium have similar densities and atomic numbers), and a stronger response to epithermal neutrons. (Indium has a large resonance at 1.4 eV; cadmium does not.) The observation that 0.1 mm of cadmium produces greater contrast than 10 times that thickness of indium (Fig. 3) shows that the neutron-radiography facility is forming an image by thermal neutrons with small epithermal-neutron or gamma-ray influence. The absence of gamma interference on the radiograph is further confirmed by noting that 2 mm of plastic material produces a higher radiographic contrast than 2 mm of lead.

A comparison of the contrast of different absorbers for each single collimator and of different collimators for each single absorber may be made. Thus the low-ratio collimator (1 : 7.5) yields much better contrast for all absorbers except lead (the gamma-radiation effect). This effect is undoubtedly due to the lower proportion of interference radiation (e.g., scattered thermal neutrons) present and helps explain why the curves of Fig. 2 do not obey the simple inverse-square law.

COLLIMATOR DIMENSIONS

The collimator ratio will be expected to affect (1) the exposure speed, (2) the geometric resolution, and (3) the relative contrasts. However, for a chosen

collimator ratio, a choice of collimator dimensions is still available. For example, a 1 : 10 collimator may be 5-mm inlet aperture and 50 mm long, or 10-mm inlet aperture and 100 mm long.

Figure 5 shows the effects of collimator dimensions on radiographic performance. Each of the three curves is for collimators with the same (1 : 15) ratio. A radiographic density of 2 can be achieved with an exposure of 15 min for the smaller 25-mm inlet aperture, whereas an exposure time of 30 min is required for the larger 50-mm inlet aperture. The smaller inlet aperture can look more precisely at the maximum of the thermal flux in the water moderator. However, the aperture should not be too small. There will be an optimum dimension, depending on the particular facility arrangement and application. For a chosen collimator ratio, as the inlet aperture dimension is decreased the collimator length will decrease, which could (1) distort the image due to wide-angle beam divergence and (2) produce excessive background interference caused by inadequate distance for shielding.

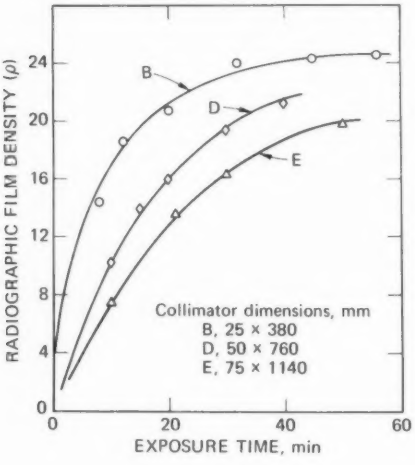

Fig. 5 ^{252}Cf neutron-radiograph transmission density as a function of exposure time for different collimator dimensions (same collimator ratio 1 : 15). Collimator input aperture immediately below source.

Figure 6 shows how the collimators compare when radiographic contrast of neutron-absorbing metals rather than overall radiographic exposure density is considered. In this case the situation is reversed—whereas the smaller dimension collimator has superior exposure characteristics, it is the larger dimension collimator that yields superior radiographic contrast

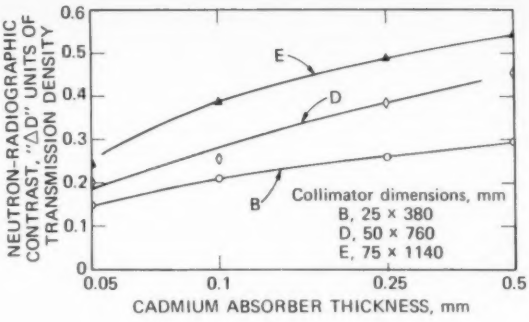

Fig. 6 ^{252}Cf neutron-radiograph contrast as a function of cadmium absorber thickness for different collimator dimensions (same collimator ratio 1 : 15).

for the neutron-absorbing materials. Neutron-flux-spectrum changes at the effective source area, together with interference radiation at the exposure position, probably account for these modifications in radiographic quality.

BEAM-EXTRACTION GEOMETRIES

The above-mentioned trials were performed with the collimator inlet aperture in a constant position as close to the source as possible without actually looking at it [Fig. 1(*b*)]. Figures 7 and 8 show that the precise point and direction of beam extraction can have very important effects on radiographic capabilities.

The density–exposure plots of Fig. 7 are for the same collimator ratio (1 : 15), the same collimator dimensions (25 by 380 mm), and the same detector combination (LiF–ZnS + Kodak Royal Blue film); the only variable is the point and direction of beam extraction. Clearly, a higher overall radiographic density is obtained by pointing the collimator directly at the source (line F). Figure 8 shows that the higher density is due to gamma radiation that comes directly from the source region rather than to increased collimated thermal-neutron intensity.

Curves B, G, and H in Fig. 7 are for collimators that look tangentially to the source to avoid unwanted gamma interference. Curve B is for the point of extraction as close as possible to the source, curve G is withdrawn 5 cm with the gap filled by water, and curve H is also withdrawn 5 cm but with the gap filled by air. Clearly the withdrawal to 5 cm, with water filling the gap, significantly reduces exposure rate (30 min for $\rho = 2$, line G, as compared with 13 min for line B). However, the withdrawal to 5 cm with air re-

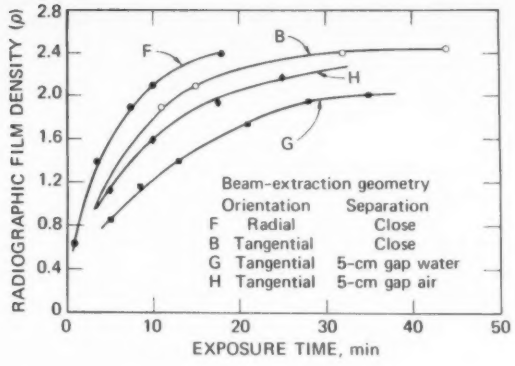

Fig. 7 ^{252}Cf neutron-radiograph film density as a function of exposure time for different beam-extraction geometries (same collimation ratio and same collimator dimensions).

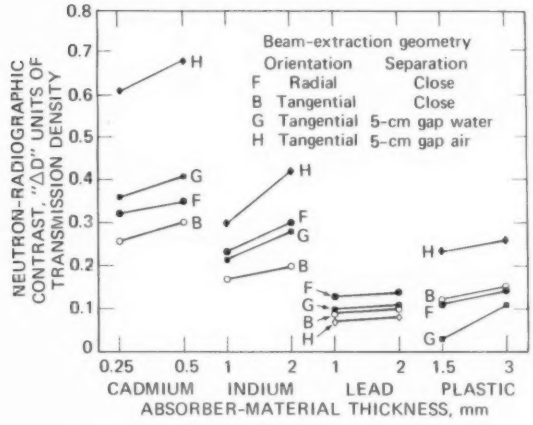

Fig. 8 ^{252}Cf neutron-radiograph contrast as a function of absorber material for four materials and for four beam-extraction geometries.

placing the water causes a much smaller reduction in exposure rate (18 min for $\rho = 2$, line H, as compared with 13 min for line B).

Figure 8, in which the radiographic contrast of four materials is plotted for each of the beam-extraction geometries, shows the significant advantage to be gained by such manipulations. The gain in exposure rate from the tangential collimator (line B) to the radial collimator (line F) is accompanied by a significant increase in lead contrast, which indicates more gamma contamination. The neutron-radiographic contrast for plastic, which is virtually gamma transparent, is not improved. The 5-cm-air-gap geometry, with the collimator line looking tangentially past the source, improves the contrasts of each of the neutron-absorbing materials (line H). The contrast is about double that of the previously standard geometry (line B). Improved neutron-energy spectrum at the effective source plane would be expected to help explain this effect.

It is additionally encouraging that, while the contrast for plastic absorber is increased in the air-gap geometry, the contrast caused by 2-mm lead is actually decreased, which indicates that there is even less gamma contamination in the radiographic image.

RESOLUTION AND OVERALL DETAIL VISIBILITY

Quantitative measurements of exposure density and radiographic contrast of selected materials are informative, but in an actual neutron-radiographic application, image resolution is of equal interest. Overall detail visibility will be a function of several interrelated factors, such as exposure density, contrast, geometric resolution, and inherent resolution.

Indicators of each of these factors are incorporated in the design of the system IQI. Inherent resolution is indicated by closely spaced holes drilled in cadmium foil positioned adjacent to the imaging plane. However, in these trials the same detection system (scintillator converter and film) was used throughout so that no changes in inherent resolution would be expected. Geometric resolution is indicated by holes in cadmium-foil strips set at increasing distances from the image plane. One set of holes is of decreasing diameter; other sets are of constant diameter and spacing. Cadmium wires of decreasing diameter set at known distances from the image plane also provide a measure of geometric resolution of high-contrast objects. A test of overall detail visibility (or low-contrast resolution) is provided by sets of 13 plastic wires placed at increasing separations from the image plane.

For the resolution tests, three observers independently read the radiographs to determine the hole and wire visibility at each separation. Each observer placed the radiographs in the same order of merit, and the results plotted in Figs. 9 and 10 are the average of the three readings.

Figure 9 shows the effect of collimator ratio on high-contrast resolution (cadmium holes and wires) and on low-contrast resolution (or overall detail visibility, i.e., plastic wires). For both high and low contrast, the

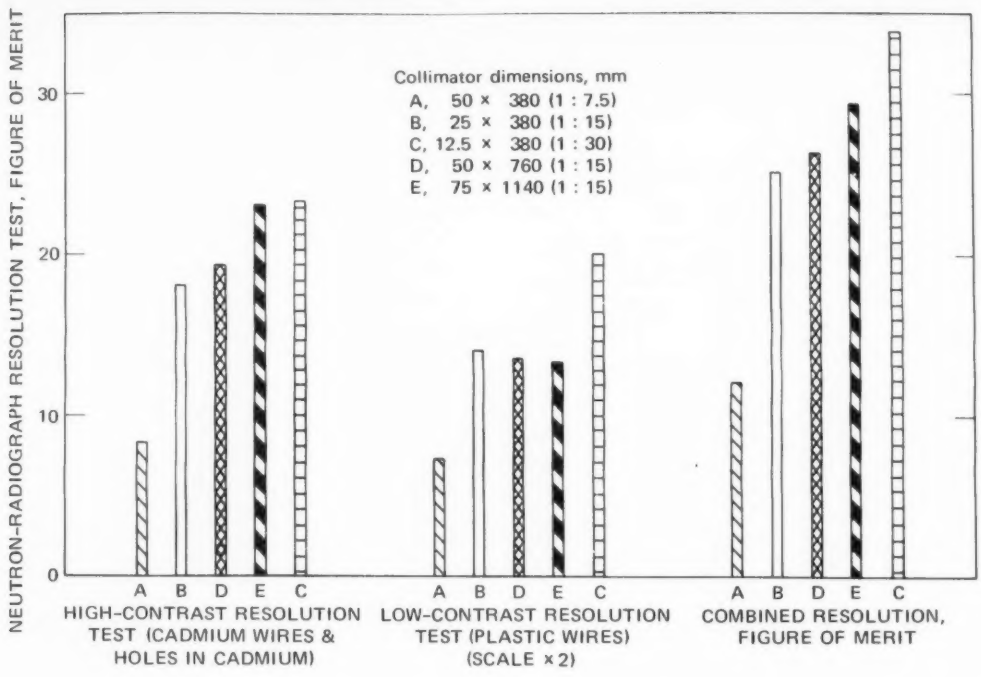
NEUTRON-RADIOGRAPH RESOLUTION TEST, FIGURE OF MERIT
Collimator dimensions, mm
A, 50 × 380 (1 : 7.5)
B, 25 × 380 (1 : 15)
C, 12.5 × 380 (1 : 30)
D, 50 × 760 (1 : 15)
E, 75 × 1140 (1 : 15)
0
10
20
30
A B D E C
HIGH-CONTRAST RESOLUTION TEST (CADMIUM WIRES & HOLES IN CADMIUM)
A B D E C
LOW-CONTRAST RESOLUTION TEST (PLASTIC WIRES) (SCALE × 2)
A B D E C
COMBINED RESOLUTION, FIGURE OF MERIT

Fig. 9 ^{252}Cf neutron-radiograph resolution figures of merit for different collimation geometries.

resolution and visibility improve rapidly with beam collimation, and the test figure of merit rises nearly linearly with collimator ratio. For a given ratio (1 : 15), the larger dimension collimator yields somewhat superior results; this can be linked with the contrast measurements discussed earlier.

The remarkably large effect of beam-extraction geometry (for a fixed collimator ratio and fixed collimator dimensions) is shown in Fig. 10. Clearly the air-gap geometry for the collimator inlet aperture is superior to the other geometries, and for this arrangement the gain is worth more than a factor-of-two increase in collimator ratio.

RADIOGRAPHIC DETECTION METHODS

It has been shown that the ratio of neutron-to-gamma radiation from a ^{252}Cf source arrangement can be sufficiently low to allow direct-exposure neutron radiography of satisfactory quality using LiF–ZnS scintillator neutron converters, and in the above-mentioned trials this method was used. A gadolinium metal-foil neutron converter is capable of providing a much finer grain image than the scintillation by the direct-exposure method. Gadolinium has a much poorer gamma-to-neutron discrimination ratio, but the experiments show that this converter may be used for neutron radiography with the ^{252}Cf source.

For neutron radiography with reactor beams, a very thin sheet of gadolinium would normally be recommended because the inherent unsharpness increases with converter-foil thickness. However, with small-source techniques we are normally concerned with applications that require lower grade images, and because of a shortage of neutrons, fairly low collimator ratios will be used. The geometric unsharpness will, in consequence, exceed the inherent unsharpness of most systems. Therefore, thicker gadolinium foils can be recommended as they are more efficient in the low-neutron fluxes and are cheaper to manufacture and easier to handle. Comparisons between 12-μ foil and 50-μ foil used with Kodak No Screen film in identical ^{252}Cf small-source arrangements show that, for equal exposure times, the 50-μ foil provides higher density and associated higher detail information. The use of double-converter screens (e.g., thin gadolinium or rhodium front screen, thick gadolinium back screen) would increase the detection efficiency.

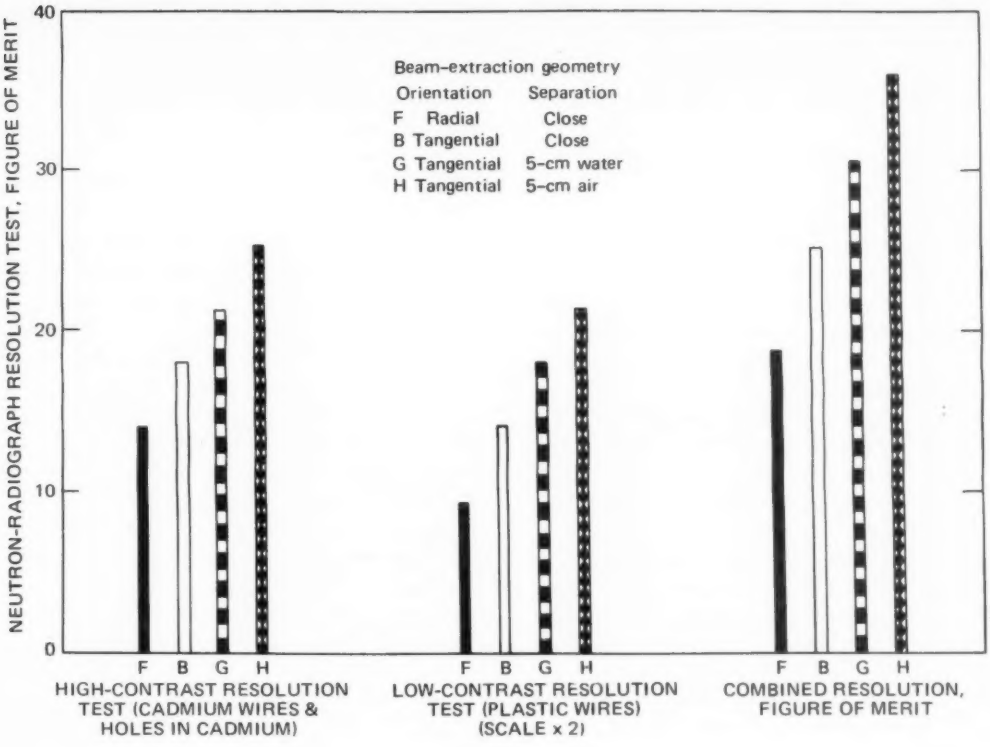

Fig. 10 ^{252}Cf neutron-radiograph resolution tests (figures of merit for different beam-extraction geometries).

The choice of film and neutron-conversion system will affect the neutron exposure required to achieve a useful radiographic density. Neutron exposures necessary to produce a film density of 1.5 for three converter-film combinations were: (1) LiF–ZnS scintillator, Royal Blue film, 7×10^5 n/cm^2 (20 min), (2) gadolinium 50-μ, No Screen film, 3.5×10^7 n/cm^2 (17 hr), and (3) gadolinium 50-μ, AA film, 1.3×10^8 n/cm^2 (64 hr). The exposure times shown in parentheses were for the 140-μg ^{252}Cf-source arrangement with a 1 : 15 collimation 7.6-cm inlet diameter and 106-cm length. The measured thermal-neutron flux at the collimator outlet was $5 \pm 2 \times 10^2$ n/(cm^2)(sec). The neutron exposures (i.e., exposure time or source intensity) differ by more than two orders of magnitude between scintillator and foil imaging methods, whereas the actual information in the radiograph is degraded only slowly in passing from the high-dose to the low-dose system. Whether to use the fine-grain gadolinium method or the faster scintillator film method must depend entirely on the requirements of the particular application.

FAST-NEUTRON INTERFERENCE

It is of interest to compare the exposures needed from small-source thermal-neutron radiography with available fast-neutron detection figures for films, converters, and point sources. When the collimator is looking directly at the ^{252}Cf source, the question arises as to whether a major or minor part of the image is due to fast neutrons coming directly from the source point.

Fast neutrons incident[9] on an X-ray film similar to Kodak No Screen or KK with no converter produce a density of 1.5 units for a fluence of 4×10^8 n/cm^2. In the thermal-neutron-radiography arrangement, a similar film density is achieved in an exposure of 17 hr for a source-to-screen distance of 100 cm. For the case where the thermal-neutron collimator looks directly at the source (3 to 4×10^8 n/sec), the direct fast-neutron exposure at the film position after 17 hr is 1.5×10^8 n/(cm^2)(sec). Thus the fast-neutron effect probably accounts for about one-third of the total exposure density.

Fast neutrons incident on film similar to Kodak Royal Blue with an X-radiography scintillator produce a density of 1.5 units for a fluence of 5×10^7 n/cm^2. Converter screens of ZnS are reported to be three times as fast for fast-neutron detection, and a density of 1.5 units would therefore be expected from a fast-neutron exposure of about 1.5×10^7 n/cm^2. In a typical thermal-neutron-radiography arrangement using Royal Blue film and ZnS scintillator, a density of 1.5 units is achieved for exposure of 12 min at a source–screen distance of 40 cm. The fast-neutron exposure in this geometry for the source of yield 3 to 4×10^8 n/sec is about 10^7 n/cm^2. Therefore, the possibility exists of a significant fast-neutron contribution to the image, and it is important to refer to IQI tests to confirm that the radiograph is basically due to thermal neutrons. The high contrast of thin sections of cadmium and the relatively low contrast of much thicker sections of lead (Fig. 8) confirm that the images are, in fact, due to thermal neutrons rather than to fast neutrons, but, as the possibility of overwhelming fast-neutron blackening does exist in some geometries, this should always be checked.

Fast-neutron exposure effects were determined for the LiF–ZnS scintillator Royal Blue film by exposing it to the ^{252}Cf with both the detector cassette and source suspended in air, separated by at least 1 m from any moderating or scattering surface. At a source–detector separation equivalent to the standard collimator B (380 mm), exposures of 5, 10, and 45 min produced film densities of 0.24, 0.53, and 1.6 units, respectively. A comparison of these figures with the thermal-exposure effects (Fig. 2) confirms that the fast neutrons contribute a significant film density.

CONCLUSION

These initial experiments were able to show that ^{252}Cf can be used for thermal-neutron radiography. The neutron-to-gamma ratio is satisfactory, and the thermalization factor of about 100 has an important advantage over other isotopic (α,n) neutron sources of the same fast-neutron yield strength. For fast-neutron radiography, the additional advantage of the small physical size of the ^{252}Cf source can be foreseen.[3]

To perform thermal-neutron radiography using small sources—isotopic sources or neutron generators—is no problem if the object is thin. In such cases, the object is placed flat against the converter–film combination, and, even if the incident thermal neutrons are virtually uncollimated, a fairly sharp image will be obtained. The challenge arises when sharp neutron-radiographic images of realistically thick objects (typically 5 mm and greater) are attempted. In this case an appropriate degree of beam collimation is required; but as the collimation is increased, difficulties of small-source neutron radiography become more pronounced.

Measurements show how the quality of neutron radiography will depend on the correct choice of factors, such as collimator ratio and beam-extraction geometry. In particular, it may be noted that (1) the inverse-square law is not followed over a range of collimator ratios from 1 : 7 to 1 : 30, (2) collimator ratio affects contrast and resolution in opposing ways, (3) collimator dimensions sensitively affect performance for a given collimator ratio, and (4) beam-extraction geometry has a large effect on radiographic capabilities, probably due to the influence of the radiation spectrum.

In practical neutron-radiography facilities based on ^{252}Cf, the source may be housed in an underground well or in an aboveground water tank to act as a convenient shield and moderator assembly. Planners should also remember the possibility of extracting several beams simultaneously from a single source.

The ultimate decision on whether ^{252}Cf can be recommended for extensive use in small-source neutron radiography (in preference to alternative sources such as Po–Be, Am–Be, Am–Cm–Be, and Pu–Be) must depend almost entirely on economic considerations, once the price of californium and the demands of particular applications can be determined.

ACKNOWLEDGMENTS

We are indebted to the AEC Savannah River Laboratory for arranging the loan of the source. Thanks are offered to the Chemistry Division at Argonne National Laboratory for providing the hot-cell facilities, and to J. A. Buzzell and T. J. Meyer of that division for help with source handling. (PSB)

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Isotope Gages: Errors in Hot-Metal-Thickness Measurement*

Summarized by Helen P. Raaen

Supplementary Keywords: *source, gamma; industry, primary metal; statistics; theory, applied; ^{137}Cs; ^{241}Am; England.*

Abstract: *Three sources of error in the measurement of the thickness of hot metals with isotope gages are considered theoretically. An artifice is proposed which may overcome some of the limitations.*

Isotope gages based on nuclear-radiation absorption are being used increasingly to measure the thickness of hot metals >5 cm thick, a use for which they are particularly suited. For relatively thick specimens, measurement errors are related especially to metal composition, temperature distribution, and statistical variation in radiation emission from the source. The article summarized here evaluates theoretically these relations for three types of stainless steel and for gamma rays from ^{137}Cs and ^{241}Am as the selected radiation sources. The evaluation method and the error magnitudes are given following a comprehensive discussion of both the basic theory of thickness measurement from nuclear-radiation penetration and the principle of constant absorption.

PRINCIPLE OF MEASUREMENT

The operation of most isotope gages is based on the equation

$$\frac{I}{I_0} = e^{-\mu\rho t}$$

where I_0 = intensity of the incident rays
I = intensity of the transmitted rays
e = 2.71828
μ = mass absorption coefficient of the metal
ρ = density of the metal
t = thickness of the metal

The term μ represents the absorption by a metal of such thickness that it has unit mass/cm^2 and is independent of the physical state of the absorbing substance. Figure 1 shows the dependence of μ on the composition of the metal (absorber) and on the energy of the radiation source. Thus, the basic assumption of constant μ is realized only under certain conditions.

ERRORS

Metal Composition

Figure 1 shows that the lower the energy level of the radiation, the more dependent μ is on metal composition, and that μ is greater for the heavier than for the lighter metals. For a radiation source whose energy is too low (e.g., ^{241}Am), an effective value of μ must be estimated for each metal whose thickness is measured. A procedure for so doing is given. In addition, the density term (ρ) of the equation is

*Summary of: F. X. Macedo and F. Seredynski, Isotope Gauges: An Analysis of Some Errors in Hot Metal Thickness Measurement, *Meas. Contr.*, **4**: T117-24 (1971).

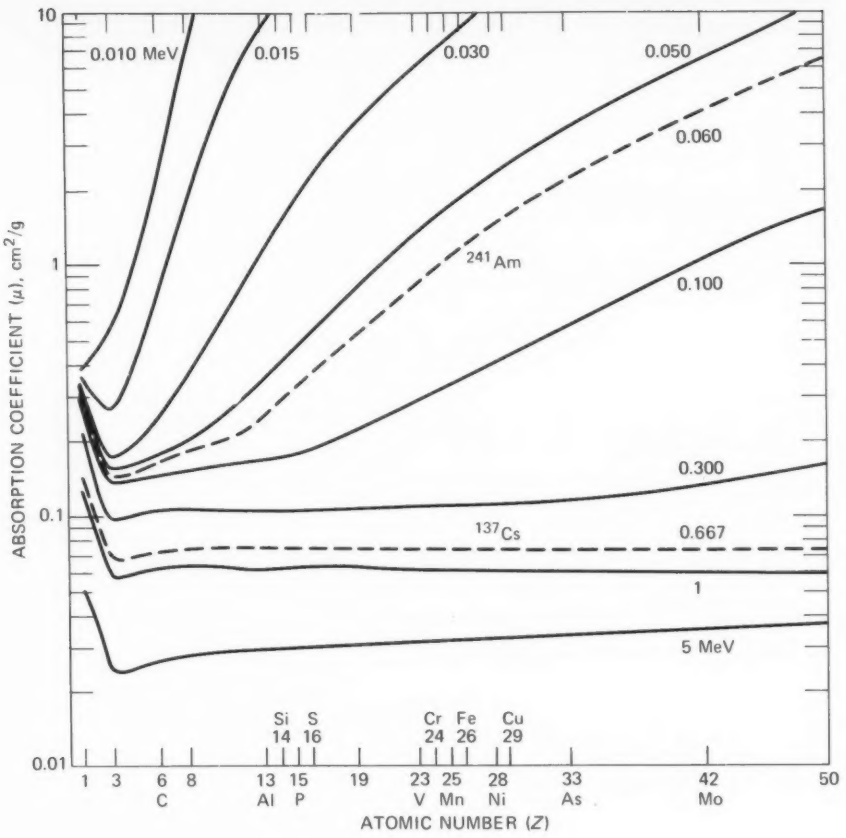

Fig. 1 Dependence of the mass absorption coefficient (μ) on the composition of the material (absorber) and on the energy of the radiation source.

composition dependent; it—as well as μ—must be compensated for when the composition of the specimen for thickness measurement varies. Metal-composition percentage-type errors for ^{137}Cs and ^{241}Am sources are estimated to be 2 and 3%, respectively.

Temperature

The term ρ is also temperature dependent. Since a constant value for a metal's coefficient of linear expansion (α) cannot be assumed beyond the temperature at which it begins to undergo a metallurgical phase change, error can be introduced in the measurement of the thickness of a hot metal. The error will be greatest within the transformation temperature range, where hysteresis effects exist. A measurement of temperature is needed to calculate ρ. A pyrometer—the means most often used to monitor the temperature of a hot metal during its irradiation for thickness measurement—cannot take into account temperature inhomogeneity (a 10% differential is possible) in the metal. For the carbon steels studied, errors resulting from the use of erroneous values of α may be about ±1.2%. The error resulting from temperature inhomogeneity through the mass of metal under study is likewise significant but decreases with decrease in metal thickness.

STATISTICAL VARIATIONS

Random variation in rate of emission of photons from a gamma source introduces a reliable (90% chance of not being exceeded) statistical error equal to $\pm 1.64\sqrt{m}$, where m is the number of particles detected. This error varies with thickness of material and with response time of the detector; it is estimated to be not less than 0.37%. Since for on-line process control,

response time must be as short as possible, the error can be decreased only by increasing the source strength—but the detector then begins to saturate.

ARTIFICE TO OVERCOME LIMITATIONS OF THE CONSTANT-ABSORPTION PRINCIPLE

The use of two isotope sources in a single isotope thickness gage is proposed to overcome limitations of the constant-absorption principle. The advantages predicted are

1. Ready increase of measuring range without loss of accuracy or sensitivity.
2. Increase in sensitivity of measurement.
3. Significant reduction, throughout the measuring range, in deviation of incident radiation intensity.
4. Increase in the ratio of minimum hot-metal thickness to maximum cold-shim thickness for any partial range.

RADIOISOTOPE-BASED MACH METER*

Summarized by Martha Gerrard

A gage for determining the ratio of the speed of aircraft relative to the speed of sound in a given atmosphere—the Mach number—has been designed by the New Radiation-Measuring Techniques Laboratory of the French Atomic Energy Commission Nuclear Research Center at Grenoble. The gage contains two ^{241}Am alpha sources and a detector for each. The long half-life (458 years) of this nuclide eliminates errors due to radioactive decay. The sources are thin platinum metal disks, approximately 10 mm diameter, in the centers of which are electrolytic deposits of ^{241}Am. The detectors are *n*-type doped silicon, approximately 1 mm thick and from a few to 30 mm in diameter.

Operation of the meter is based on the difference in pressure between the inner and outer surfaces of the aircraft. If the pressure on the outer surface (impact pressure) is designated by p_i and that on the inner surface (static pressure) by p_s, then

$$p_i - p_s = \rho V^2/2$$

where ρ is the density of the atmosphere surrounding the aircraft and V is the rate of the craft relative to the atmosphere. The value of p_i is assumed to be greater than that of p_s. The attenuation of the radiation in the two chambers depends on the pressure of the gas in them, and thus the detector readings can be converted to a Mach number (Fig. 1).

The instrument can be adapted to operate as a combined altimeter, altitude stabilizer, and Mach meter.

*Summary of Mach Meter Using Semiconductor Radiation Detectors for Nuclear Radiation (in French), French Report CEA-R-4107, by Louis Gilly, Paul Jourdan, and Pierre Denis, January 1971.

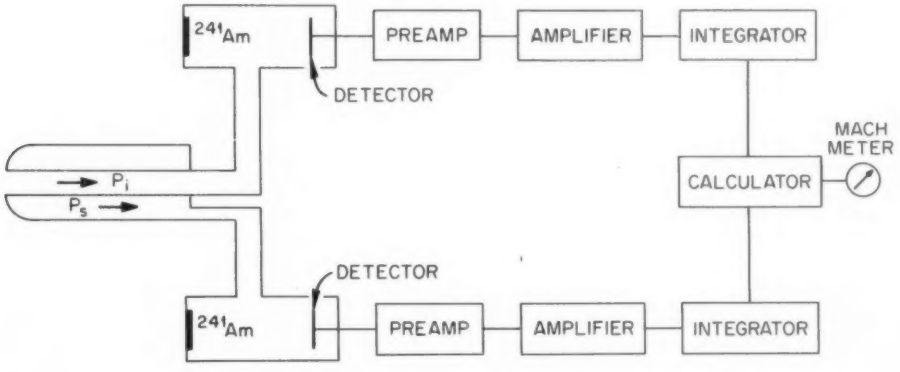

Fig. 1 Diagram of gage for determining Mach number.

PROGRAM TO EVALUATE THE EDUCATIONAL USES OF A ^{252}Cf SOURCE IN A NUCLEAR SCIENCE AND ENGINEERING PROGRAM*

A program has been carried out at the Georgia Institute of Technology under a Division of Isotopes Development contract to evaluate the potential usefulness of ^{252}Cf sources in college and university courses. The source used was 119 μg of ^{252}Cf, made at Savannah River.

The source was shipped in a cylindrical container (Fig. 1) consisting of a lead-shielded concentric pipe inside a 55-gal drum filled with paraffin. A wax plug in the tubular source holder prevented neutron

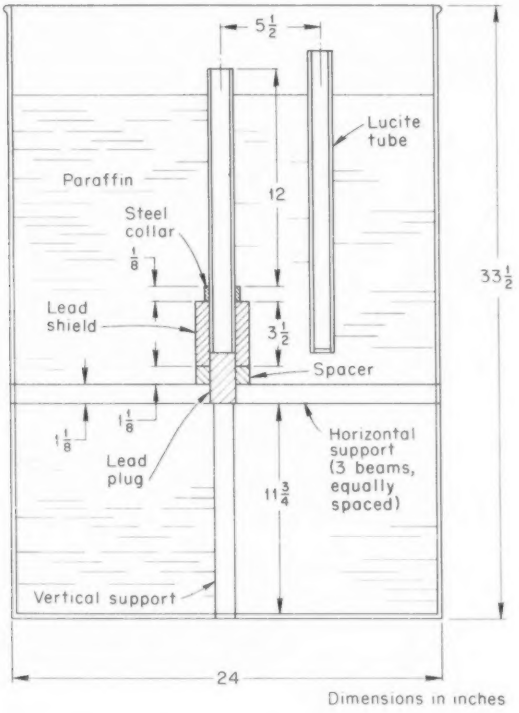

Fig. 1 Source storage container.

streaming. Upon receipt of the source at Georgia Tech, a vertical Lucite tube was installed parallel to the source holder to permit activation of small samples without moving the source. In most of the studies, the source was kept in the container. Among the experiments considered suitable for course work were: determination of boron in borosilicate glass; measurement of Fermi age in water; activation analysis of industrial effluents for manganese, copper, and nickel; gamma radiography of metal samples; fission and alpha-particle etching of plastics; and irradiation of ethyl iodide for Szilard–Chalmers separation. (MG)

*Abstract of Program to Evaluate the Educational Uses of a ^{252}Cf Source in a Nuclear Science and Engineering Program, Annual Report, January–December 1970, USAEC Report TID-25752, 1971, by G. G. Eichholz, School of Nuclear Engineering, Georgia Institute of Technology, Atlanta.

Environmental Radiation Surveys and Snow-Mass Predictions from Aircraft

By L. J. Deal, J. F. Doyle, Z. G. Burson, and A. E. Fritzsche*

(Summarized by Martha Gerrard)

Supplementary Keywords: *hydrology; measurement, moisture;* ^{40}K; ^{208}Tl.

Abstract. *A system is described for determining the water equivalent of the snow cover by using the natural radioactivity of the soil and airborne radiation detectors. This is a summary of USAEC Report EGG-1183-1522, Technical Report L-1034, June 25, 1971.*

For 10 years a sophisticated aerial radiation detection and tracking system—the Aerial Radiological Measuring System (ARMS)—has been in operation to assist the U. S. Atomic Energy Commission in monitoring the impact of nuclear facilities on their environment. It provides both real-time and passive records of nuclear radiation levels from isotopes that might be in the air, deposited on the ground over a large area as point sources, or contained in the soil as naturally occurring isotopes.

The aerial platform is a Twin Beach Bonanza Model E-50, which has a survey capability of slightly more than 3 hr at a flying speed of 150 knots. The primary instrumentation consists of fourteen 4- by 4-in. NaI(Tl) scintillation crystals, pulse-height analyzers, Doppler radar positioning equipment, and paper-tape recording equipment.[1,2]

The system has been used to document the natural and man-made radiation levels on the ground around 45 nuclear sites and facilities covering 60 thousand square miles; search for a lost radioactive source,[3] a nuclear submarine, a SNAP power source, and strayed missiles;[4] and to define and track effluents produced from power-reactor operations, nuclear rocket tests, and Plowshare cratering events. The system "stands by" (in the air) with an alpha spectrometer for all manned Apollo missions.

*EG&G, Inc., 680 E. Sunset Road, Las Vegas, Nev. 89101 (L. J. Deal is now with the USAEC Division of Biology and Medicine, Washington, D. C.). This work (Report EGG-1183-1522) was reported at the 7th International Symposium on Remote Sensing of Environment, Ann Arbor, Mich., May 17–21, 1971. Some subsequent data are given in EGG-1183-1533, Technical Report No. L-1047, Oct. 22, 1971, Water Equivalent of Snow Measurements Using Natural Terrain Radiation, by A. E. Fritzsche, Z. G. Burson, and D. L. Burge.

An interesting practical application of radiation surveys made from the air is determination of the water equivalent of the snow cover by measuring the attenuation of natural radiation through the snow to obtain data for estimating the water equivalent of snow for use in water-resources prediction and flood control. Current means of estimating water equivalent are generally limited to individual points, and the unevenness of snow deposits necessitates many separate measurements to obtain meaningful estimates for large watersheds. Feasibility studies based on this technique have been reported from Russia,[5–7] Norway,[8] and the United States.[9]

This article summarizes the first major field tests[10,11] with the ARMS system over snow courses in the United States—near Luverne, Minn., and Steamboat Springs, Colo.—during late 1969 and the first 8 months of 1970. The work was sponsored by the National Oceanic and Atmospheric Administration. Extensive measurements were made by the standard snow-tube method to correlate with the airborne data for all surveys.

DETERMINATION OF WATER EQUIVALENT OF SNOW

In measuring the gamma attenuation through the snow cover, the gamma flux (from ^{40}K, ^{208}Tl, etc.), $\psi(E)$, at altitude h observed by an airborne detector over snow-covered ground, at water equivalent depth d, containing a monoenergetic isotope of concentration A in the soil, is given by

$$\psi(E) = \frac{A}{2\,\mu_g(E)} \int_1^\infty \frac{e^{-xt}}{t^2}\,dt$$

where t = sec θ (angle between perpendicular and a ground point) and $x = \mu_a(h + 1.1d)$. The symbols μ_a and μ_g are the mass attenuation coefficients for a gamma ray of energy E for air and soil, respectively. The coefficient for water is a factor of 1.11 greater than that for air over the range of energies of interest.

The attenuation vs. air and/or water depth reduces to the ratio of two exponential integrals for the uncollided component. The equations for total photons arriving at the detector must include all scattering events and have no simple closed analytic representation like the equation above but must be solved by Monte Carlo calculations. Attenuation curves vs. water depth for ^{40}K and ^{208}Tl (a 3.1-min radionuclide naturally occurring in the ^{228}Th decay scheme) and an estimated net count (gross count less background) are given in Fig. 1.

The basic technique for correlating gamma intensity with snow mass is to measure the intensity first for bare earth and then to measure for as many different snow-mass thicknesses as is practical. Intensity data were acquired at several altitudes—from 200 to 1000 ft—above the ground at both the Luverne and Steamboat Springs snow courses, and the gross-count data were recorded vs. position every second (equivalent to 200 ft of ground path) in flight. The spectrum data were obtained over about 4 miles of flight and represent an average over that distance.

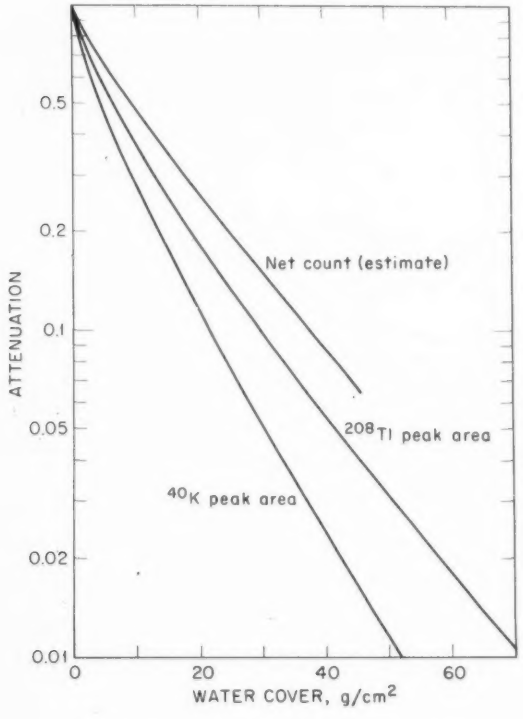

Fig. 1 Gamma attenuation as a function of water depth (theoretical).

The gross-count and spectrum data were treated separately. The known aircraft plus cosmic-ray background in the gross count was removed, and the net count was then averaged over each mile of snow course. The peak areas due to ^{208}Tl (2.62 MeV) and ^{40}K (1.46 MeV) were extracted from the spectrum data. Both net count and peak areas were plotted against snow mass, air mass, and a combination of the two, and, from these plots, the effect of both snow and air mass on gamma attenuation was evaluated. Gamma attenuation due to snow cover is expected to be proportional to that due to air since the gamma interaction cross sections are proportional for all energies of interest.

A sample of the net count vs. water depth is shown in Fig. 2. Even with no corrections for soil moisture or

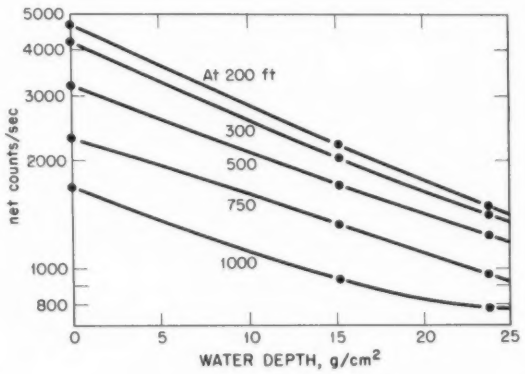

Fig. 2 Net count as a function of water depth. Points, from left to right, represent values determined on May 13, Apr. 16, and Mar. 14, 1970, respectively.

air-density changes, the curves are smooth and values decrease as expected. Displacement of the curves is caused by changes in soil moisture (vertical) and snow cover (horizontal). At higher altitudes the data are strongly affected by airborne radon, which was not corrected for. The net count vs. a combination of water and air-mass thickness follows a well-defined curve (Fig. 3). Spread in the data may be due to the difficulties of background subtraction.

An example of peak area analysis (uncollided gamma component) to determine water depth is shown in Fig. 4. Again, the displacement in the curves is due primarily to snow cover.

The natural-terrain radiation levels over dry ground vary from point to point by as much as a factor of 2. Normalization to the "no-snow" condition then be-

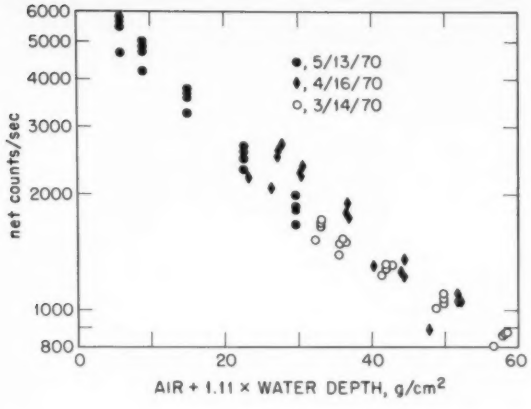

Fig. 3 Net count as a function of air-layer thickness plus water depth.

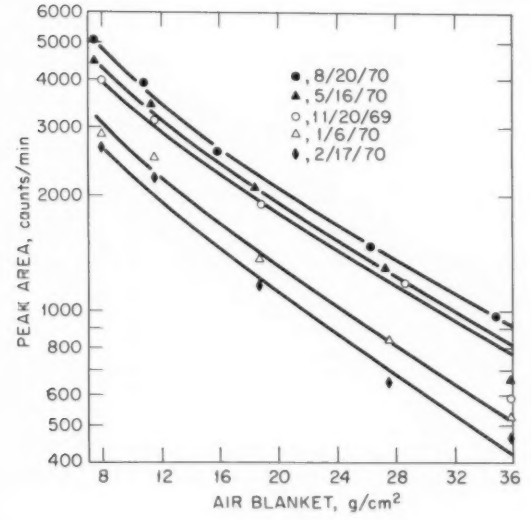

Fig. 4 Peak area count as a function of air-blanket thickness.

comes necessary for maximum accuracy in prediction of water equivalent from snow cover.

These data from the first field trials, taken with equipment designed for other tasks and not optimized for snow surveys, were very encouraging. Equivalent-water predictions were obtained for up to 10 in. of water cover. Standard deviations from averaged snow-tube data were 23% for peak area data and 17% for gross-count data taken at low altitudes. However, snow-tube data also contain some uncertainties, and it is expected that, with better definitions of airborne radon, cosmic radiation, and soil-moisture effects, the uncertainties will be less.

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None of the references here have been verified by the editors.

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AREA MEASUREMENT WITH A RADIOACTIVE PREPARATION*

The surface area of a thin, flat plate may be measured by interposing it between a planar radioactive source and a large-window detector. In the system that was tested (Fig. 1), a 300-mCi ^{241}Am alpha source of 16.5 cm diameter was used with a counter having a 20-cm-diameter window (0.9 mg/cm^2). The area to be measured had to be smaller than the 13.8-cm-diameter opening in the sample holder, this opening being covered with 10-μ-thick plastic. The count rate recorded by the system electronics depended linearly on the surface area under study (Fig. 2). (MG)

*Abstract of article (in German) by H. Eggelsmann and W. Rasp, *PTB Mitteilungen*, **80**: 436-439 (1970).

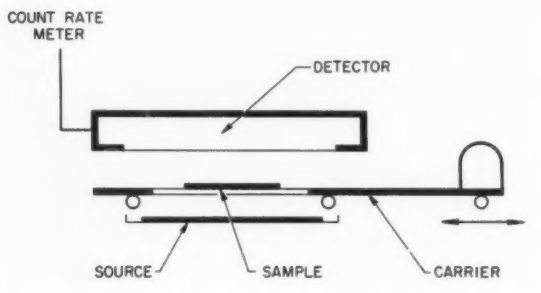

Fig. 1 Surface-area measuring system.

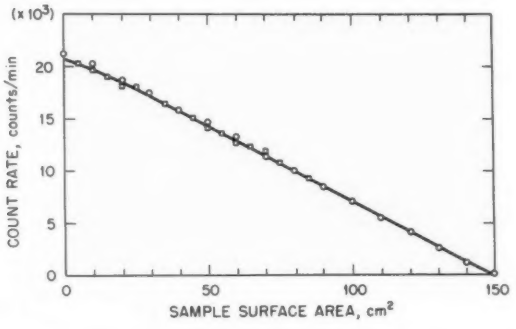

Fig. 2 Dependence of alpha count rate on surface area of (○) circular samples and (□) square samples.

RADIOISOTOPE DEPTH INDICATOR*

For measuring the depth which a logging cable has reached at any moment, ^{90}Sr–^{90}Y is deposited on the cable. As the cable moves downward, the radioactivity is continuously detected by six G–M counters arranged in a circle around the cable. The measurement is accurate within 6 mm at cable speeds up to 2000 m/hr. (MG)

*Abstract of Automatic Device for Monitoring Radioactive Depth Marks of Logging Cables (in Czech) by A. Zboril and S. Mares, *Geologicky Pruzkum*, **12**(3): 82-84 (March 1970).

PASSIVE DEVICE FOR DETERMINING RELATIVE ROTATION*

An instrument for determining relative rotation of a body has a radioactive inert gas—^{85}Kr—retained in a definite pattern in the lattice of the material of an inertial element. Detectors are positioned on the equator and at the end of the axis of the unit (Fig. 1) so as to sense the radioactive and nonradioactive areas of the source pattern. The detector outputs are determined by the displacement of the inertial element. The isotopic sensor has the advantage that it does not add a load to the gyroscope and thereby cause precessing. (MG)

*Abstract of U. S. Patent 3,531,997, Oct. 6, 1970 (filed June 30, 1967), W. C. Eddy, Jr., and J. P. Scheiwe, Assigned to Industrial Nucleonics Corporation.

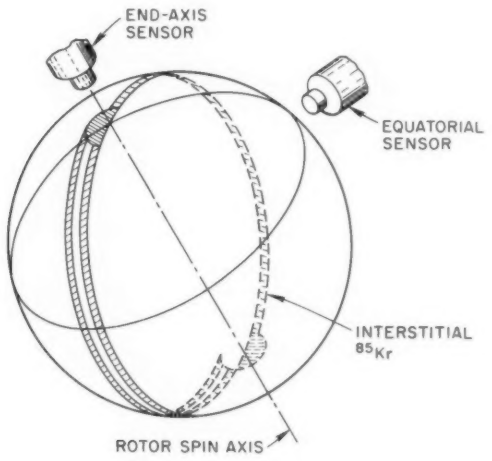

Fig. 1 Spherical gyro system with radioactive source at poles and in pole-to-pole strip.

NUCLEONIC MEASURING APPARATUS WITH AUTOMATIC FIRE SAFETY RADIATION SOURCE SHUTTER CLOSING AND LOCKING MEANS CONTROLLED BY A FUSIBLE ELEMENT WHICH MELTS AT HIGH TEMPERATURES*

This invention relates to a safety device for a nuclear gage (e.g., thickness or density measurement) which automatically shields the source in the event of fire or high ambient temperature. A unit such as an electric solenoid moves a heat-resistant shutter, which, in the closed position, effectively blocks or shields the radiation emanating from the radiation source. Normally, during operation of the gage, the shutter is maintained in the open position by continuous energizing of a solenoid or by an air-operated motor. A temperature-responsive element, such as a fuse, upon reaching a predetermined temperature, causes the shutter to close and seal the source capsule within its housing, rendering it harmless in the event of a fire. At a second, still higher, predetermined temperature, a spring-loaded pin in a retracted position and retained by a solder barrier is released when the solder barrier melts and effectively locks the shutter in the closed position, ensuring protection within the housing. (REG)

*R. E. Bach (assigned to Industrial Nucleonics Corporation), U. S. Patent 3,567,942, Mar. 2, 1971 (filed Apr. 27, 1967).

MEASUREMENT OF MOTOR OPERATING TEMPERATURES USING ^{85}Kr*

Preliminary tests indicated tnat the temperature at which a motor has been operating can be determined by the radio-release technique using ^{85}Kr. First, ^{85}Kr is entrapped in a metal part, e.g., a piston ring, which is installed in the motor. After the motor has been operated for a given length of time, it is shut down and allowed to cool, the part is removed, and the residual ^{85}Kr is measured. Experiments made both with metal pieces heated to known temperatures and with metal parts installed in motors showed that, for a given heating time, the amount of ^{85}Kr remaining in the metal was a linear function of the temperature to which the metal had been heated (Fig. 1). The maximum temperature at which the linear relation holds differed for different metals. (MG)

*Abstract of article (in German) by J. F. Ury in *Isotopenpraxis*, **7**: 21-27 (1971).

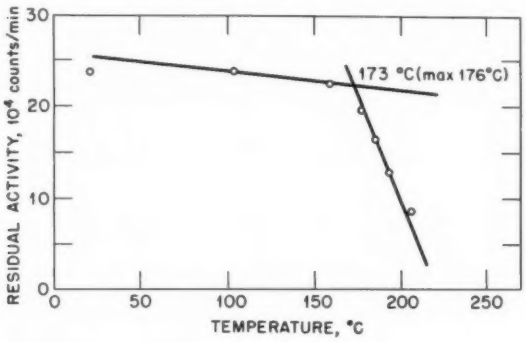

Fig. 1 Residual ^{85}Kr activity in an aluminum strip after being heated to various temperatures.

Solid-State Diffusion: Familiar Examples and Modern Problems

By J. W. Kelly*

Supplementary Keywords: *activable tracer.*

Abstract: *Techniques for detection of solid-state diffusion, including the use of activable tracers, are discussed.*

Diffusion—the phenomenon that occurs when atoms of the same or different materials intermingle—is usually considered to occur at extremely low rates in solids. However, many familiar processes in solids depend on diffusion occurring at appreciable rates[1]—the rusting of steel by direct oxidation, the sintering process used to produce uranium dioxide fuel for atomic reactors, the diffusion of alloying elements, and the diffusion of minority carriers across the base region from emitter to collector junctions in semiconductors. There is evidence that atoms in solids have a residence time of less than 1 μsec. Thus there is a paradox—macroscopic evidence, such as the stability of mountains, suggests atoms to be immobile, while atomic evidence suggests the opposite. The resolution of this

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paradox, together with pressure from modern-day technology that demands ever-improving properties from the materials it uses, has led to the recent upsurge of interest in solid-state diffusion, a subject that has been investigated continually for nearly a century.

DIFFUSION COEFFICIENTS

The rate of diffusion is given by the diffusion coefficient, D, from Fick's First Law, which states that the flux, J_A, of a component A diffusing in the x direction is proportional to the concentration gradient,

$$J_A = -D^{dC_A}/dx$$

at constant temperature (the negative sign indicates that matter is transported from the higher to the lower concentration region). D is measured in square centimeters per second, which corresponds to the diffusing material penetrating up to 0.001 in. in 1 min and 10,000 years, respectively.

Prior to 1945, most diffusion experiments were carried out with two chemical species in appreciable concentrations,[1a] D being here the "chemical diffusion coefficient" and, contrary to the ideas then prevailing, a function of the concentrations and self-diffusion coefficients of both species. The wide variety of artificial radioisotopes that have become available since then has enabled diffusion studies to be carried out with very low (impurity diffusion) and zero (self-diffusion) chemical–concentration–gradient conditions. The results from these latter types of studies are more readily compared with theoretical models of diffusion than are those from chemical-diffusion experiments and so have tended to replace them.

In the most popular method[1b] for experimental determination of D, a thin layer of radioactive material is deposited on one end of the sample and allowed to diffuse in at a constant temperature. The value D is found from the concentration–penetration profile obtained by postdiffusion sectioning. This method is capable of precision within a few percent and may be used to measure values of D ranging from 10^{-7} cm^2/sec for carbon in steel at 1000°C to 10^{-12} cm^2/sec for beryllium in alumina at 1150°C. At the AAEC Research Establishment, Lucas Heights, it has been used to measure the rate of diffusion of beryllium in both alumina and beryllia.

For ionic solids, a voltage is placed across the material and the current that flows is measured. The mobility, μ—i.e., the diffusion—of the ions of charge q is responsible for the electric current and is measured by determining the electrical conductivity of the solid and the diffusion coefficient. From the Nernst–Einstein equation

$$D/\mu = kT/q$$

where k is Boltzmann's constant and T the temperature.

However, the standard techniques are inapplicable if there are no satisfactory isotopes, if the values of D to be measured are lower than 10^{-12} to 10^{-13} cm^2/sec, if several ions may be contributing to the electrical conductivity, etc.

Measurement of the rate of diffusion of oxygen in ceramic oxides is a typical example. Of the four radioactive oxygen isotopes, ^{15}O has the longest half-life, 2.1 min, which is much too short for the standard sectioning technique to be applicable. Hence, recently developed methods for such measurements are based on the detection of stable ^{18}O (or ^{17}O), which has a low natural abundance compared with ^{16}O. In the mass spectrometer method,[2] the sample is heated in an atmosphere enriched in ^{18}O and a mass spectrometer is used to monitor the rate at which ^{18}O exchanges with ^{16}O in the sample. In the activation-analysis method,[3] after the diffusion anneal in an ^{18}O-enriched atmosphere, a shallow taper is ground on the surface of the sample to reveal the ^{18}O gradient. The sample is then irradiated in a high-energy accelerator, producing the ^{18}O$(p,n)^{18}$F nuclear reaction, and D is determined from an autoradiograph of the ^{18}F isotope distribution. At Lucas Heights, this method has been used to measure the diffusion rate of ^{18}O in BeO.

For the measurement of very low diffusion rates in metals ($D = 10^{-16}$ to 10^{-20} cm^2/sec), the inelastic relaxation method[4] has been most used. It is restricted to binary alloy systems in which stress-induced solute ordering occurs. The relaxation time, τ, for internal friction due to such solute rearrangements is determined, and D is calculated from

$$D = a^2/36\tau$$

where a is the lattice parameter. The method is well established for interstitial solid solutions (Snoek relaxation), but difficulties in theoretical interpretation limit its use with substitutional alloys (Zener relaxation).

This method is indirect in that a property dependent on mass transfer, rather than mass transfer itself, is measured, and only in the last decade have direct methods, using radioisotopes and nonsectioning techniques, become available for the measurement of low

values of D. In Savitsky's method,[5] introduced in 1963, the metal (or nonmetal) sample is annealed in a molten salt bath of the same metal spiked with a suitable isotope, and D is determined from the exchange of activity with the sample. Savitsky used silver and silver nitrate to measure a value of $D = 10^{-19}$ cm^2/sec for self-diffusion in silver at 255°C. The method is analogous to the mass-spectrometer method for oxygen.

Methods based on the absorption of radiation have also become available. In a technique due to Gainotti and Zecchina,[6] a thin layer of radioactive tracer is deposited on one surface of a thin foil, and the intensity of radiation emitted from the sample is measured initially [$I(0)$] and after a time t [$I(t)$]; D is determined from

$$\frac{I(t)}{I(0)} = 1 + 2\mu\left(\frac{Dt}{\pi}\right)^{1/2} + \mu^2 Dt$$

where μ is the radiation absorption coefficient. If the radiation spectrum is complex, an absorption method[7] in which the intensity is measured on both sides (I_1 and I_2) of the foil as a function of time, D can be determined from

$$\ln\left(\frac{I_1 - I_2}{I_1 + I_2}\right) = \ln k - \frac{\pi^2 D}{x^2}$$

where x is the thickness of the sample and k is a constant. The author has used this method at Lucas Heights to measure the rate of self-diffusion in alpha zirconium at 550°C when $D = 10^{-16}$ cm^2/sec.

DIFFUSION MECHANISMS

The discovery that gold diffuses more rapidly in lead at 300°C than does the chloride ion from common salt in water at 15°C led Schottky and Frenkel, in 1930, to make the greatest single step in our understanding of diffusion mechanisms. They advanced the then radical idea that solids were not perfect crystals but contained defects, and diffusion occurred as a result of atoms jumping from one lattice site to the next via the defect. Basically, two types of defects have been proposed: a vacancy in which an atom is missing from the lattice [Fig. 1(*a*)] and an interstitial in which an extra atom is included in a nonlattice position [Figs. 1(*b*) to (*d*)]. Nondefect mechanisms involving 2- or 4-ring atom exchanges have also been proposed [Figs. 1(*e*) and (*f*)].

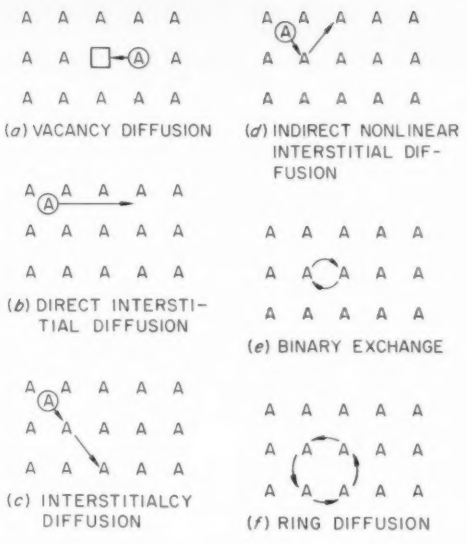

Fig. 1 Suggested mechanisms of diffusion.

The activation energy for diffusion, Q, was then seen as the energy barrier for atoms jumping via these defects and could be calculated theoretically for the simpler cubic lattices. Typical results[8] from such calculations for copper are

Defect	Q, kcal/mole
Interstitial	97
Binary exchange	240
4-ring exchange	90
Vacancy	45

The experimental value of 47 kcal/mole for Q indicates a vacancy mechanism, and this is true in general for metals. In ionic salts, the condition of electrical neutrality demands that both cations and anions take part in the diffusion process. When the sizes of the ions are similar, as with NaCl, the defects are thought to be cation and anion vacancies (Schottky defects); when they are appreciably different, as with AgCl, the defects are thought to be equal numbers of interstitial silver ions and silver vacancies (Frenkel defects).

Kirkendall Effect

Not until 1947 was there found strong evidence supporting a vacancy-diffusion process in metals and alloys. Smigelskas and Kirkendall[9] showed that if inert

molybdenum markers were placed at the initial boundary between a 30% brass and copper diffusion couple, diffusion annealing moved them from this position (the Kirkendall effect), and porosity (holes) developed on one side of the interface. Since marker displacements can occur only if the copper and zinc atoms move at different rates, exchange mechanisms that prohibit such displacements are eliminated and a vacancy mechanism becomes the most probable process (interstitial diffusion is most unlikely in this system).

Since then the Kirkendall effect has been found in many cubic-metal-alloy systems, in oxide and sulfide films, and in a few interdiffusing salt pairs. Thus diffusion by a *defect* process is now well established, and examples of exchange mechanisms, if they occur at all, will have to be sought in the many noncubic structures still to be examined.

Correlation Factor (*f*)

The fact that it took 17 years for strong experimental evidence to be found for vacancy diffusion in metal systems points to the difficulty of this kind of study and to the desirability of a direct and simpler means of identification. Bardeen and Herring[10] opened the way for such a development in 1952 by pointing out that, in vacancy diffusion, an atom does not jump from one position to the next in a fully random manner. For example, in Fig. 1(*a*), if atom A on the right-hand side of the vacancy jumps into the vacancy position, it will more probably revert to its initial position on the next jump than move to any other position. Thus successive atom jumps are not random, they are correlated. This correlation decreases the overall diffusion by the factor f, so that high degrees of correlation correspond to small values of f.

The value of f is determined purely by the geometry of the lattice and the jump mechanism and has been calculated, using random walk theory, for a number of structures and mechanisms:

Defect	Lattice	f (calculated)
Vacancy	Simple cubic	0.65
Vacancy	Face-centered cubic	0.78
Vacancy	Body-centered cubic	0.73
Direct interstitial	Face-centered cubic	1
Interstitialcy	Face-centered cubic	0.33
Indirect nonlinear interstitial	Face-centered cubic	0.73

The experimental method by which f could be measured was suggested in 1958 by Schoen[11] and by Tharmalingam and Lidiard,[12] who independently proposed the equation

$$1-\frac{D_\beta}{D_\alpha}=f\left[1-\left(\frac{m_\alpha}{m_\beta}\right)^{1/2}\right]$$

The D's and m's are the diffusion coefficients and masses of two isotopes, α and β, of the same element. Since m_α and m_β are known, direct evaluation of f is reduced to an accurate measurement of D_α and D_β. Results for some systems for which satisfactory isotopes are available are

Isotopes	Matrix	f (measured)	Diffusion mechanism
$^{22}Na/^{24}Na$	NaCl	0.78	Vacancy
$^{22}Na/^{24}Na$	Na	0.36	Interstitialcy
$^{65}Zn/^{69}Zn$	Zn	0.73	Vacancy
$^{6}Li/^{7}Li$	Si	0.94	Direct interstitial
$^{12}C/^{13}C$	Fe	1.07	Interstitial
$^{103}Pd/^{112}Pd$	Pd	0.81	Vacancy

Recent refinements of correlation theory have shown that the method can be used not only to identify the diffusion mechanism but also to provide details of the jump process itself, such as the fraction of the translational kinetic energy carried through the saddle-point configuration by the jumping atom.

Other scientifically interesting aspects of modern solid-state diffusion studies, not discussed here, are the unusual non-Arrhenius behavior of beta zirconium, the vacancy wind effect, and the near-surface diffusion anomaly in which refined sectioning techniques have shown diffusion to occur at a higher rate adjacent to the surface than in the bulk.

Thermal Diffusion

In the above discussions it has been assumed that diffusion occurs at a fixed temperature because most diffusion studies have been of this type. Only in the last decade have extended measurements been made of solid-state diffusion in a temperature gradient, i.e., thermal diffusion or the Soret effect. Although this effect was first noted in metals in 1926 by Ballay,[13]

who observed a 0.6% variation in lead content between the 200 and 300°C regions of an initially uniform, heated lead–thallium alloy, the difficulties of such experiments largely discouraged further work until recently.

The introduction of a temperature gradient modifies Fick's First Law, which becomes

$$J = -D\left(\frac{\partial G}{\partial x} + \frac{Q^* G}{RT^2}\cdot\frac{dT}{dx}\right)$$

where R is the universal gas constant and Q^*, the experimentally derived "heat of transport," describes both the magnitude and sign of the thermal-diffusion effect. When it is negative, the diffusing species migrates up the temperature gradient so that, contrary to what is expected intuitively, the diffusing species concentrates in the hot end. A good example of this is the diffusion of carbon in steel. If Q is positive, diffusion occurs down the temperature gradient as in the diffusion of hydrogen in zirconium.

This latter example has important implications in power reactors using zirconium alloys for pressure tubes. Hydrogen from the decomposition of the cooling water dissolves in the zirconium, and when the solubility limit is exceeded (~400 ppM at 390°C), platelets of zirconium hydride form and embrittle the zirconium. To avoid this, designers have been forced to use thicker tubes, and metallurgists have sought to minimize the deleterious effects of hydride precipitation by controlling the orientation of the precipitates.

In 1966 Wilkins and Sawatzky[14] pointed out that the use of the Soret effect provided a simple and elegant solution to the problem. They attached zirconium cooling fins to a zirconium pressure tube to establish a temperature gradient between the cool unstressed fin and the hot stressed pressure tube. They then added 500 ppM of hydrogen to both fin and tube, so that hydride was present at the start of the experiment, and annealed the finned tube at a mean temperature of 390°C. Within 100 days, the hydrogen concentration in the tube decreased to 250 ppM, below the solubility limit, and rose to 2000 ppM in the fin. Thus they demonstrated that, using the Soret effect, hydrogen could be moved from a harmful position in the stressed pressure tube, where an amount in excess of the solubility limit could cause failure, to a harmless position in the unstressed fin.

The importance of thermal diffusion is not restricted to power reactors. The subject is also of vital concern in areas of modern technology where metals are required to have high strength at high temperatures and in temperature gradients—for example, the turbine blades of jet aircraft. The strength of many materials depends on the formation and stability of finely dispersed precipitates having a well-defined size range. When these precipitates grow beyond a critical size or dissolve in the matrix, the strength is impaired. Hence, if such problems and others are to be avoided, and if older alloys are to be improved and new ones developed, there is a clear need for further studies both of isothermal diffusion and the experimentally and theoretically more difficult thermal diffusion.

(MG)

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IMPROVEMENTS IN OR RELATING TO PROTECTING SECURITIES AGAINST FORGERY*

This invention concerns improvements in protecting securities against forgery and is specifically designed to be applied to any security as a certification marking which indicates that the security is valid for some specific purpose. The marking is an area of radiation-sensitive printing ink or paint, and certification is validated by a change in color of the ink or paint upon electron-beam irradiation. Two classes of ink or paint may be used: (1) a dye sensitive to radiation and dispersed in an inert carrier or (2) a composite of a radiation-sensitive carrier and a dye that detects changes in the carrier. An example of the first class is methylene blue, which is bleached by radiation, in a cellulose derivative. Dyes that develop a color upon irradiation may also be used. Class 2 indicators are preferred. These incorporate a halogen-containing polymer—for example, poly(vinyl chloride) or poly(vinylidene chloride)—which is sensitive to radiation and an indicator that changes color in the presence of hydrogen chloride (which is liberated upon radiolysis of the polymer by an electron beam). A protective coating may be placed over the radiation-sensitive area so that the developed marking is not inadvertently destroyed by the ultraviolet component of daylight. (REG)

*F. L. Dalton and J. D. McCann, British Patent No. 1,193,511 (assigned to United Kingdom Atomic Energy Authority), June 3, 1970 (filed June 21, 1967).

Measurement of Gas Distribution with ^{41}Ar

By C. L. Williams, J. H. Rust, and G. T. Colwell*

Supplementary Keywords: *tracer; material tracing; diffusion; pollution; atmosphere.*

Abstract: *The use of ^{41}Ar to measure the distribution of a gas in a container is described. The equipment, procedure, and results are discussed.*

The relative or absolute distribution of a gas in a container can be measured remotely by monitoring the radioactivity from a gamma-emitting component gas. The feasibility of this technique was demonstrated in experiments performed to provide confidence in future research on measuring gas distributions in operating heat pipes that contain noncondensable gases. Argon-41 was the radioactive gas. This technique may also be used to measure concentration profiles in the steady-state jet mixing of two dissimilar gases and to test the effectiveness of a gas collector in antipollution devices.

EQUIPMENT

The equipment (Fig. 1) consisted of an ^{41}Ar-supply capsule, a test section of known volume distribution, and radiation-detecting instrumentation. The test section was a 1.50-cm-ID steel tube that contained an axially centered aluminum cone (40.6 cm long by 1.36 cm maximum diameter). This arrangement gave a parabolic volume distribution for the gas in the test section. Irradiated argon was used as the gas because the ^{41}Ar half-life (110 min) is short enough to prevent safety problems following a 24-hr period after a test and because its gamma energy (1.29 MeV) is sufficiently large that the photopeak is detected above background radiation. A 5.08- x 5.08-cm NaI(Tl) crystal mounted on a multiplier phototube was used to detect the gamma radiation from ^{41}Ar. The multiplier-phototube output was analyzed with a TMC 400-channel multichannel analyzer. The crystal and multiplier phototube were enclosed by a 10-cm-thick lead shield, with the exception of a 1.27- x 1.78-cm opening, which served as a collimator to permit gamma radiation from the corresponding location in the steel

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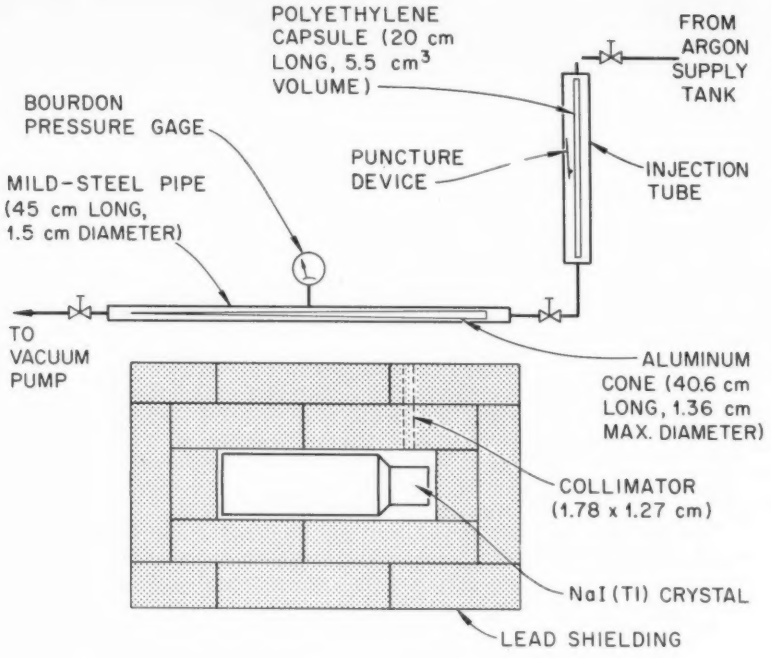

POLYETHYLENE CAPSULE (20 cm LONG, 5.5 cm³ VOLUME)
FROM ARGON SUPPLY TANK
BOURDON PRESSURE GAGE
PUNCTURE DEVICE
INJECTION TUBE
MILD-STEEL PIPE (45 cm LONG, 1.5 cm DIAMETER)
TO VACUUM PUMP
ALUMINUM CONE (40.6 cm LONG, 1.36 cm MAX. DIAMETER)
COLLIMATOR (1.78 x 1.27 cm)
NaI(Tl) CRYSTAL
LEAD SHIELDING

Fig. 1 Schematic diagram of the equipment.

tube to enter the crystal. The shielded assembly was mounted on a track so that gamma scans could be made along the entire length of the steel tube.

PROCEDURE

To produce ^{41}Ar, a 5.5-cm^3 polyethylene capsule filled with argon (at 2 atm pressure) was irradiated for 8 hr in the Georgia Institute of Technology Research Reactor; ~110 mCi of ^{41}Ar was produced. The capsule was placed in the injection tube, and the entire system was evacuated. Argon-41 was introduced into the test section by puncturing the capsule and bleeding in the irradiated argon to a desired fill pressure. To reduce background and Compton scattered gamma rays from the lead shield, the gamma spectrum was measured in the region of the ^{41}Ar 1.29-MeV photopeak; measurements were made at eight fixed axial locations along the test section and were repeated at the same positions with the collimator closed. Background and ^{41}Ar-decay corrections were made to the data. The difference between the count rates with the collimator open and closed was assumed to be due to ^{41}Ar radioactivity in a 1.27-cm-long volume along the axis of the test section.

RESULTS

Figure 2 shows the results; normalized void volume is plotted as a function of axial position. The normalized void volume is the gas volume over the length along which measurements were taken; it is also given

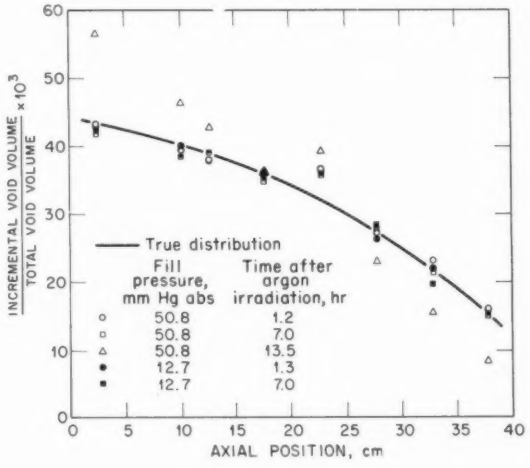


Fig. 2 Results of gas-distribution measurements.

by the count rate (at some axial position) divided by the integral of the count rate over the length along which measurements were taken. For the measurements taken 1.2 and 7 hr after argon irradiation, the measured void distributions were within 5% of the calculated values. Measurements taken 13.5 hr after argon irradiation show considerable scatter, which is due to statistical errors caused by the low count rates. The high readings at position 22.9 cm resulted from the extra volume of ^{41}Ar present in the Bourdon pressure gage. Errors in the measurements caused by attenuation of the 1.29-MeV ^{41}Ar gamma ray are estimated to be <5% at the thick end of the aluminum cone.

The results show that gas distributions can be measured accurately by monitoring ^{41}Ar gamma radiation. However, because statistical errors increase with decrease in ^{41}Ar radioactivity, the technique is limited to use not later than ~7 hr after neutron irradiation of the argon. (HPR)

SHORT-LIVED COSMOGENIC RADIONUCLIDES TRACE IN-CLOUD SCAVENGING PROCESSES*

Supplementary Keywords: *hydrology; meteorology; material tracing; in situ tracer; kinetics; reaction mechanism; spallation; ion exchange; spectrometry; counting, coincidence; atmosphere; mobile; review, technical; ^{24}Na; ^{38}Cl; ^{39}Cl.*

Cosmic-ray bombardment of the atmosphere produces cosmogenic radionuclides at a relatively steady rate. Because these radionuclides rapidly become attached to the natural aerosol, they can trace its subsequent behavior. To define rates and mechanisms of in-cloud processes, the more abundant short-lived cosmogenic radionuclides ^{38}Cl (37.3 min), ^{39}Cl (55 min), and ^{24}Na (15 hr) were measured rapidly and consecutively during three rainstorms.

The experimental procedures were interesting. The rain-collection area was a 450-m^2 plastic-covered surface on the roof of an aircraft hangar at the Quillayute, Wash., airfield. The collected rainwater drained from the plastic sheet through a plastic hose into a large drum, from which it was pumped through a water sampler that included beds of anion- and cation-exchange resins, which retained the ^{38}Cl and ^{39}Cl and the ^{24}Na, respectively. At the rain-collection site, the ^{38}Cl and ^{39}Cl were determined by coincidence counting with two mobile multidimensional gamma-ray spectrometers; the counting precision was better than ±10%. The resin samples that contained the longer-lived ^{24}Na were counted for longer periods in a laboratory under conditions that gave the same precision.

The observations provided information about the sequence of in-cloud rainwater development. The cosmogenic radionuclides ^{24}Na, ^{38}Cl, and ^{39}Cl serve as condensation nuclei for cloud-droplet formation. As the cloud droplets grow further by coalescence and condensation, the additional collection of newly formed cosmogenic radionuclides is small or negligible. The in-cloud raindrops, especially in light rains, may undergo several cycles of partial evaporation followed by additional coalescence and condensation. Light rains develop more slowly than heavy rains and have higher radionuclide concentrations and higher ratios of long- to short-lived radionuclides. A 30- to 130-min period is required for raindrop development in light to medium rains. In the rainstorms studied, the apparent time since previous rainout had cleaned the air ranged from 30 to 200 min—an estimate that needs verification.

The natural cosmogenic radionuclides were shown to be extremely useful as tracers for studying the rates and mechanisms of in-cloud scavenging processes. The studies indicated the need for better supporting meteorological information—for example, cloud thickness, local and large-scale atmospheric motions, size and electrical charge of raindrops, and weather radar. (HPR)

*Abstract of R. W. Perkins, C. W. Thomas, and J. A. Young, Application of Short-Lived Cosmogenic Radionuclides as Tracers of In-Cloud Scavenging Processes, *Journal of Geophysical Research*, 75(15): 3076-3087 (1970).

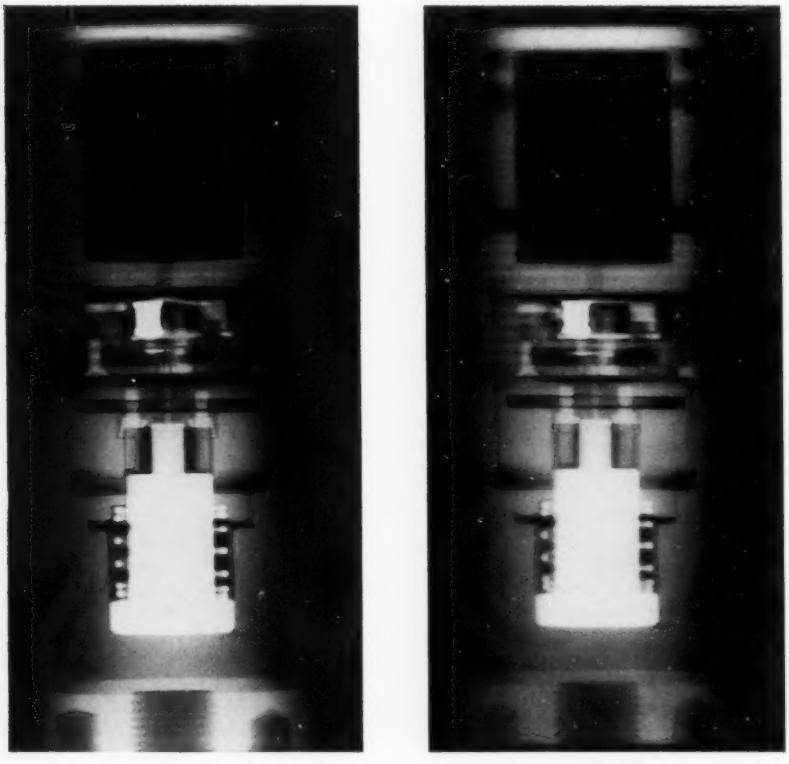
USE OF ^{169}Yb IN INDUSTRIAL RADIOGRAPHY*

The generally available radioisotopes used for gamma radiography emit gamma radiation of energy too high for use in inspection of thin ferrous or light-alloy specimens. Radiographs made with a source produced by activating natural ytterbium to produce a mixture of ^{169}Yb and ^{175}Yb—which emits gammas ranging from 0.11 to 0.4 MeV—were compared with X radiographs to determine the usefulness of this nuclide in industrial radiography.

The gamma radiographs made with a 1-Ci ^{169}Yb source compared favorably with X radiographs (Fig. 1). Exposure curves had been made with stepped wedges in preliminary experiments, and defects in thin metal specimens, not detectable with ^{192}Ir radiation, had been demonstrated with ^{169}Yb.

Because of its short half-life (~31 days) and the low curie strengths available (4 to 6 Ci with a 2-mm-diameter source), ^{169}Yb is not a real competitor of ^{192}Ir for radiography in the thickness range for which ^{192}Ir is an acceptable source. However, a ^{169}Yb source could probably be used on steel thicknesses down to about 5 mm and on aluminum and its alloys down to about 10 mm with better sensitivity to flaws than can be obtained with ^{192}Ir. Exposure times can be shorter than with a ^{170}Tm source of the same physical size, and, unlike ^{170}Tm, ^{169}Yb has no bremsstrahlung. (MG)

*Summary of An Assessment of ^{169}Yb for Industrial Radiography, R. Halmshaw, T. J. Manly, and M. Dowd, *British Journal of Non-Destructive Testing*, **13**(4): 102-105 (1971).



(a) (b)

Fig. 1 Prints of radiographs of ordnance fuse: (*a*) ^{169}Yb radiograph and (*b*) X radiograph, 250-kV X rays, heavily filtered. British Crown copyright reserved.

Bureau Eurisotop

In November 1961 there was created, within the general Department of Industry and Economy of the European Community of Atomic Energy, a service responsible for the promotion of the use of isotopes and radiation in economic, technical, and industrial areas. This service was designated Bureau Eurisotop. The activities of the Bureau, under the direction of Dr. G. Pröpstl, encompass the following broad responsibilities.

1. *Focusing on applications, methodology, and equipment.* Development work is not carried out in Euratom laboratories but is done under contract through cooperation of Euratom with industrial organizations and scientific institutions of the European Community. Euratom contributes both financing and personnel for these endeavors.

2. *Providing information and documentation.* The Bureau maintains a diversified collection of, as well as a bibliography of, publications relating to the use of isotopes and radiation. This service is ordinarily provided without cost to organizations, firms, and persons of the Community.

In addition, the Bureau Eurisotop publishes, in collaboration with experts of the European Community, information brochures, in which the material is classified by areas, methods, and principles of applications. These include bibliographies, proceedings of meetings and work sessions, case studies, and state-of-the-art monographs, which are published in one or more languages, including French, German, Dutch, and Italian.

3. *Coordinating efforts in the European Community.* By sponsoring work sessions dealing with special topics, the Bureau tries to improve the overall efficiency of the numerous individual efforts directed toward isotope applications. In these sessions, there are examined the problems of allocation of work areas, establishment of an overall research and development program, and promotion of isotope applications.

4. *Answering legal and economic questions.* With the multiplicity of possibilities of isotope and radiation applications, there are many legal and economic questions that are not sufficiently well recognized. The Bureau studies these problems and reports on them.

To date, a number of booklets have been published covering specific areas within the scope of *Isotopes and Radiation Technology.* Brief résumés of these are included below.

These documents, if in stock, can be obtained from

Bureau Eurisotop–Euratom
Rue de la Loi, 200
1040 Brussels, Belgium (PSB)

GENERAL

Selected Bibliography of Euratom Reports Dealing with the Utilization of Radioisotopes, Eurisotop Booklet No. 17, Bibliographic Series No. 4, 78 pp., 1966.

This particular booklet constitutes a "first try" at providing a general look at Euratom publications concerned with technology of the applications of radioisotopes and radiation. For each of ~230 items, there are included the title, author(s), report number, and journal or other publication reference where applicable, plus an abstract in the original language and in English. Unfortunately, there is no index and there seems to be no particular organization to the abstracts. However, they are quite informative. (PSB)

Isotopes and Radiation in Industry and Technology (in French, German, Italian, or Dutch), Eurisotop Booklet No. 3, Monograph Series No. 1, 96 pp., June 1965.

These booklets present a series of papers that were given at a symposium in Brussels sponsored by Eurisotop and the Bank of Brussels on the industrial and technical aspects of radioisotopes and radiation. The following papers are included:

1. Manufacturing Artificial Radioisotopes, by R. Constant.

2. The Electronics Industry and the Use of Radioisotopes, by J. Perette.

3. Use of Radionuclides in the Iron and Steel Industry, by A. Kohn.

4. The Importance of Radioactive Material for Research and Technology in the Chemical and Pharmaceutical Industries, by H. Götte.

5. Radioisotopes in the Textile Industry, by M. Robinet.

6. Activation Analysis—Theory and Use, by J. Hoste.

7. Nuclear Energy, Agriculture, and Food Industry, by F. Van Hoeck.

8. Problems Presented by the Application of Radioactive Tracers in Hydrology, by J. N. Svasek.

9. Radioisotopes in Internal Medicine, by L. Heilmeger.

10. Use of Radioisotopes and Radiation in the Machine Industry, by C. P. Galotte.

11. Present and Future Research Problems in the Field of Industrial Use of Radioisotopes, by R. Cornuet.

All the articles are written at a very elementary level and should be easily understood by anyone who has even a small technical background. Most of the articles do not have bibliographies. (RHL)

Conference on Radiation and Isotope Techniques in Civil Engineering—Abstracts of Papers (in French, German, Italian, Dutch, or English), Eurisotop Booklet No. 51, Bibliographic Series No. 6, Part 1, 55 pp., August 1970; Part 2, 20 pp., September 1970.

These booklets contain abstracts of 56 papers that were presented at a Conference on Radiation and Isotope Techniques in Civil Engineering held at Brussels, Oct. 28–29, 1970. The highlights of this conference were recently summarized by G. J. Rotariu and M. Steinberg in *Isotopes and Radiation Technology*, 9(1): 5-8 (Fall 1971).

The conference held sessions on the following subjects: Mineral-Based Material Combinations by Radiation Chemistry (concrete–polymers); Hydrology; Industrial Production of Construction Elements and Materials; Control and Regulation on Construction Site; Earthworks, Foundations, Traffic Engineering, Soil Testing; Material Combinations Based on Wood and Natural Fibers by Radiation Chemistry (wood–plastic composites); Testing of Building Materials (nondestructive property measurements); Protection of Buildings, Utilities Ventilation (fire protection, lightning conductors, air conditioning, and underground leak detection); Environment of Civil Engineering Structures (waste-water treatment, dust emission from stacks); Testing of Civil Engineering Structures (leak detection, gamma-ray radiography of concrete and mortar joints); Hydraulic Engineering (coastal protection, hydraulic transport, dam construction); Radiation Protection in Civil Engineering (construction of shielding walls, natural radioactivity of construction materials, isotope transportation containers).

While many conferences on isotope applications have been held over the past 15 years, this conference was the first international one devoted specifically to the building industry and civil engineering. When the full proceedings of the conference are published, they will make a valuable addition to the literature of isotope applications. (RHL)

IRAD Activities To Encourage the Use of Irradiation Techniques, Eurisotop Booklets: No. 26, F. Busse (Ed.), Bremerhaven symposium, Sept. 24–27, 1967 (in German), 85 pp.; No. 27, Kiel symposium, Sept. 30–Oct. 10, 1967 (in German), 67 pp.; No. 28, W. Koeck and B. Ording (Eds.), Duesseldorf symposium, Oct. 9–15, 1967 (in German), 116 pp.; No. 29, O. van Wauwe (Ed.), Kortrijk symposium, Oct. 18–22, 1967 (in Dutch), 100 pp.; No. 30, R. Mulkens (Ed.), Charleroi symposium, Oct. 24–29, 1967 (in French), 151 pp.; No. 39, C. Rispal (Ed.), Lyon symposium, May 3–12, 1968 (in French), 193 pp.; No. 40, J. F. Diehl (Ed.), Munich symposium, May 18–26, 1968 (in German), 134 pp.; Conference Proceedings Series 5, 6, 7, 8, 9, 12, 13.

The Eurisotop organization, with its project designated "Aktion IRAD," is sponsoring 10 symposiums in the European Community, 7 of which are reported in the booklets mentioned above. Papers presented at these meetings, and printed in the booklets, are general reviews on large-scale uses of radiation—e.g., sterilization of medical products; polymerization, coating treatment, and wood–polymer manufacture; food preservation; mutation production and male insect sterilization; and uses of radiation in the textile industry. A few facility descriptions are presented, and economics, legal problems, and the future of irradiation technology are considered. (MG)

Technical–Economic Study of Automatic Mandrel-Break Safety Device with a Radioisotope in the Cold-Pilgering of Seamless Tubes (in German or Italian), Eurisotop Booklet No. 14, Case Study Series No. 4, Berthold and coworkers, 2nd edition, 13 pp., 1969.

This booklet describes a safety device that applies gamma radiation from ^{137}Cs to rapidly detect breakage of the mandrels used in the rolling step of the cold-pilgering process to make seamless metal tubes. The device has considerable economic significance—savings into the millions of dollars. It increases production by decreasing the time the machine stands while ingots to be formed into the tubes are loaded. By automatically and rapidly shutting down the machine when a mandrel breaks, it prevents costly downtime and repairs.

The bulletin consists in the sections

1. The Cold-Rolling of Tubes.

2. The Importance of the Mandrel-Break Safety Device.

3. Mechanical Mandrel-Break Safety Device.

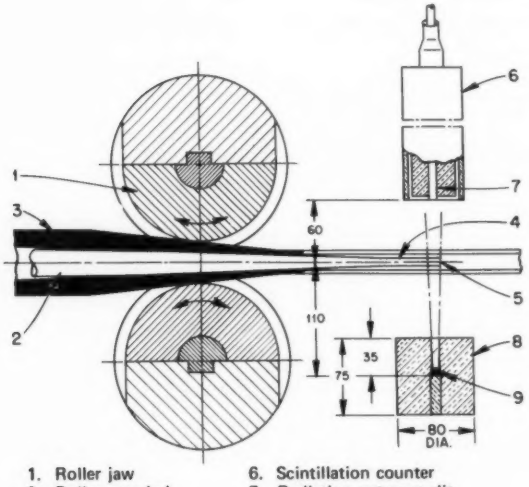
4. Mandrel-Break Safety Device by Means of a

Radioactive Source.

5. Technical Comparison of Methods.
6. Economic Comparison of Methods.
7. Work-Training Aspects and Motivation.
8. Economic Survey.

The first section gives a brief history of the cold-pilger process, reviews the advantages the process has over older processes, especially with regard to greater ease of handling the ingots and to achieving better uniformity in dimensions of the product, and the recent increase in the use of the process as the result of the construction of fast-running and multiple cold-pilger machines. The second section explains how the high-speed multiple operations increase the chance for mandrel breaks—on the average, two breaks per day in work with 15 machines—and thus also increase the need for a safety device that acts as quickly as possible to detect a break and to automatically stop the machine. The very brief third section describes a mechanical safety device, used on the older machines, whereby rods, fastened on the mandrel and extending over the whole length of the fabricated tube, are used to detect deviate travel of the mandrel and to activate a switch that shuts down the machine. The mechanical device, however, is too slow for the newer machines. The newer device (Fig. 1) is described in detail in section 4. Information is given about the ^{137}Cs gamma-radiation source and its shielding, the NaI-type detector, and the count-rate meter. By suitable choice of range of measurement for the radiation impulse, the device is adaptable over the entire range of tube diameters and wall thicknesses encountered in the rolling process. A relay circuit, regulated by the summed output current of the count-rate meter acts on the drive of the machine with such sensitivity that proper ingot feed rate and sufficiently rapid detection of a mandrel break are always ensured. The large number of these devices in use (67 on June 1, 1965) indicates their satisfactory technical performance, except for minor troubles associated with vibration. Section 5 presents the technical advantages of the radioisotopic safety device compared with the mechanical device. These include better control of ingot feed, mandrel motion, and machine shutdown. The determinable economic advantages are discussed in section 6—decrease in "still time" of the machine, reduction in number of employees, greater reliability of machine performance, elimination of damage to the machine assembly by a broken mandrel, and overall increase in production. The results of an economic survey indicated a net saving of about $8000. The problems described in section 7 relate to work training and motivation in the application of the radioisotopic device. They have to do primarily with personnel difficulties associated with fear of radiation—an unnecessary fear that stemmed, in two interesting but regrettable cases cited (a blood-donation program and an objection to health-physics regulations), from lack of adequate and proper information and from mishandling of labor relations.

The closing note is significant—" . . . Meantime, all the remaining cold-pilger machines in the world are rapidly being equipped with mandrel-break safety devices that are based on the described use of radioactive radiation." (HPR)



1. Roller jaw
2. Roller mandrel
3. Tube ingot
4. Rolled tube
5. Elongated rod filled at the front with lead
6. Scintillation counter
7. Radiation entrance slit
8. Lead shield
9. Radioactive rays, 6 mCi ^{137}Cs

Fig. 1 Pilger cold-rolling machine equipped with a radioisotopic mandrel-breakage safety device.

GAGING

Level Gaging

Nine booklets describe the use of radioisotopic level gages to measure and control the level of various materials in process equipment. The most noteworthy fact in these nine studies is the wide variety of materials whose levels are measured. These range from such very-low-density materials as acetaldehyde and polyethylene to such very-high-density materials as

iron ore and molten steel. The levels of both liquids and powders are measured. Each study contains data on economics of using the gage as well as a technical description of the gage and its installation.

Radiometric Level Gauging in Power Plants (in German), H. Wieczorek and H. Erhardt, Eurisotop Booklet No. 4, Case Study No. 1, 13 pp., August 1965.

Radiometric Level Gauging in the Production of Mineral Wool (in German or Italian), H. Wieczorek and H. Erhardt, Eurisotop Booklet No. 10, Case Study No. 2, 11 pp., September 1965.

Radiometric Level Gauging in a Thermal Cracking Plant (in German or Italian), H. Wieczorek and H. Erhardt, Eurisotop Booklet No. 13, Case Study No. 3, 6 pp., October 1965.

Radiometric Level Gauging in the Production of Alumina (in German or Italian), H. Wieczorek and H. Erhardt, Eurisotop Booklet No. 32, Case Study No. 6, 25 pp., March 1969.

Measuring Charge Levels in Cupola Furnaces Using Radioisotopes (in German or Italian), Eurisotop Booklet No. 37, Case Study No. 7, 19 pp., December 1969.

Radiometric Level Gauging in the Production of Low-Pressure Polyethylene (in German or Italian), Eurisotop Booklet No. 46, Case Study No. 9, 12 pp., May 1970.

Radiometric Level Gauging in the Production of High-Pressure Polyethylene (in German), Eurisotop Booklet No. 47, Case Study No. 10, 16 pp., August 1970.

Radiometric Level Gauging and Control in Vertical Continuous Casting Plants for Steel (in German), Eurisotop Booklet No. 48, Case Study No. 13, August 1970.

Radiometric Level Gauging in Production of Acetaldehyde (in German), Eurisotop Booklet No. 49, Case Study No. 11, August 1970. (RHL)

Moisture Gaging

Moisture Measurement with Neutrons (in French and German), A. Trost, Eurisotop Booklet No. 12, Monograph Series No. 3, 46 pp., 1966 (23 references).

This booklet reviews briefly the conventional methods of moisture measurement and then, after a consideration of the principles of the nuclear method and the effects of various parameters (e.g., density, composition, and size of sample), neutron sources, sondes, and detector amplification are discussed. The neutron source of choice is ^{241}Am–Be.

The applications covered include glass sand, foundry sand, sinter mixtures, coal and coke, production of concrete and calcareous sandstone, and soil studies (including surveys of building sites, agricultural applications, and hydrological and geological measurements).

The booklet concludes with a brief section on radiation protection and a bibliography. (PSB)

Radiometric Moisture Determination in the Preparation of Ready-Mix Concrete (in German or Italian), Berthold, Eurisotop Booklet No. 38, Case Study Series No. 8, 21 pp., 1969.

After an introduction that outlines some of the details of the production of cement and discusses the importance of the measurement of the exact moisture content of components added to the cement mixture, the author mentions the conventional methods of moisture analysis and then considers the nuclear technique for moisture determination. The measuring sonde, containing an ^{241}Am–Be neutron source that provides 3×10^5 fast neutrons/sec, and the auxiliary equipment are described. The sonde also contains the detector for slow neutrons. The device is suitable for continuous measurement of sand. Technical and economic considerations (as of 1964) conclude the presentation. (PSB)

Thickness Gaging

Thickness Measurements in a Paper Mill by Beta Attenuation (in Italian), E. E. Petrozzi, Eurisotop Booklet No. 50, Case Study Series No. 14, 27 pp., 1970.

Continuous measurement of the thickness of a moving sheet of paper is described. The radiation from a beta-emitting radioisotope—e.g., ^{147}Pm, ^{204}Tl, or ^{90}Sr–^{90}Y—passes through the paper to a detector, with attenuation a function of paper thickness; variations in the thickness are indicated by variations in the count rate. Two types of support are mentioned: (1) a "C" frame, with the source in one arm and the detector in the other, and (2) a "bridge" frame, which has a transverse bar above the paper and another below, with the source attached to one bar and the detector to the other so that they move simultaneously across the width of the paper as the paper goes forward longitudinally. (MG)

Density Gaging

Radioisotopic Measurements of Crude Oil Density in Pipelines (in German, French, or Italian), H. Wieczorek and H. Erhardt, Eurisotop Booklet No. 31, Case Study Series No. 5, 14 pp., 1968.

The density of crude oil flowing in a pipeline was measured continuously by determining the attenuation of the gamma radiation from a 2-Ci ^{137}Cs source as the radiation passed through the pipeline (Fig. 1). A

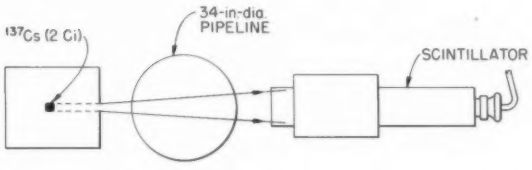


Fig. 1 Apparatus for continuous measurement of crude-oil density in a pipeline.

scintillation counter was used for the measurement. With Libyan oil, density 0.84 g/ml, the counter reading was 10,000 counts/sec; with Savannia oil, density 0.87 g/ml, the reading was 2500 counts/sec. The recording (Fig. 2) clearly showed the 0.03 g/ml difference. Thus it was possible to make a sharp cutoff when changing from one oil to another. (MG)

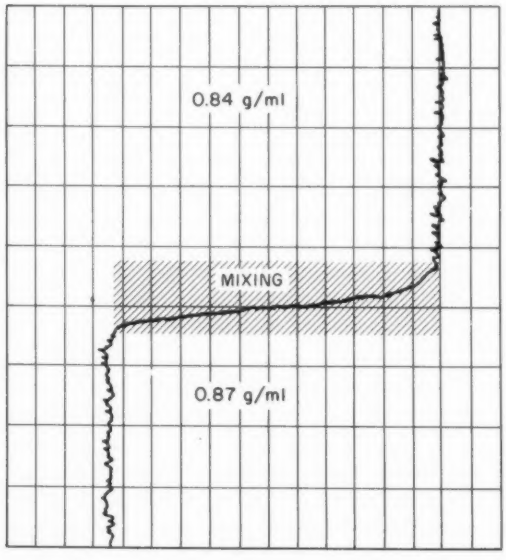


Fig. 2 Change in count-rate record with change in crude-oil density of 0.03 g/ml.

TRACERS

Use of Radioactive Tracers for Studying Gas Leaks (in Italian or German), Ferdinando Borelli and Pier Giorgio Caldera, Eurisotop Booklet No. 15, Reports Series No. 4, Summary of Proceedings of Symposium at Turin, May 21, 1964, 35 pp., 1966.

The feasibility of using radioisotope-labeled preparations—e.g., $CH_3{}^{82}Br$—for detecting leaks in gas-conducting piping was discussed, and methods for injecting tracers were demonstrated. Photographs and equipment sketches and diagrams are included, but there are no references. (MG)

Tracer Methods for Measuring Solids Movement in Water (in German or French), Proceedings of Conference in Brussels, Oct. 2–4, 1963, Eurisotop Booklet No. 7, Vol. 1, papers and discussions, 156 pp.; Eurisotop Booklet No. 8, Vol. 2, complete papers, 489 pp., Conference Proceedings Series Nos. 1 and 2, 1965.

Twenty-nine papers were presented at the conference, most of which reported work with radioactive tracers but a few with fluorescent tracers. Natural sand and silt, as well as glass-simulating sand grains, were labeled with various radioisotopes—^{198}Au, ^{51}Cr, ^{182}Ta, ^{170}Tm, ^{192}Ir, ^{46}Sc, ^{95}Zr, ^{144}Ce, ^{140}Ba, and others. Subjects covered include solids transport in water and along river bottoms, dune and channel formation, methods for labeling natural and artificial sediments, experimental techniques, safety measures required, and recommendations for further study. Emphasis is on studies in the European Community. (MG)

Autoradiography (in French), Eurisotop Booklet No. 34, Monograph Series No. 9, by A. Kohn, 62 pp., 1969.

In recognition of the unique position of autoradiography among methods that use isotopes and radiation, the author of *Autoradiography* (Eurisotop 34) has two purposes: to outline the various phases of the technique of autoradiography and to compare its use in the two chief, but widely different, fields of application—metallurgy and biology. These purposes are accomplished in a concise and factual presentation.

The book is introduced with a brief summary of the special features of autoradiography, the procedure for making an autoradiograph, and its various uses. References to general reviews on its use in biology and metallurgy are cited. Three chapters follow: I, Theoretical Considerations on Autoradiography; II, Practical Conditions for the Execution of Autoradiographs; and

III, Some Examples of the Application of Autoradiography. An appendix defines and discusses the "coefficient of autoradiographic efficiency."

The theoretical considerations include image formation and resolving power of the autoradiographic image and cover (1) the characteristics of the photographic emulsion and the photochemical changes induced in the emulsion by the radionuclide contained in the material being autoradiographed and (2) the influence of emulsion–sample contact, thickness of emulsion and of sample, and energy of radiation on resolving power. Practical factors that affect resolution are also discussed, including kind of emitted radiation (beta, X, gamma), energy of emitted beta radiation (the kind most commonly used), grain size and thickness of the photographic emulsion, and the significance of the distance between the sample and the emulsion.

The two principal techniques for obtaining autoradiographs—contact (particularly applicable to metals) and track (especially for biological samples)—are presented separately in detail; the quantitation of autoradiographs is then discussed. Color autoradiography, comparison of autoradiographic with microradiographic images, and autoradiography effected in a reactor are given special attention. Also, photographs of the arrangement for a 30-ton ingot show that the contact procedure for autoradiographing samples of many square meters in area is feasible. The preparation of samples, e.g., histological specimens, handling of emulsions (especially stripping emulsions), autoradiographic exposure, and examination of track autoradiographs in an electron microscope are all carefully summarized.

The final chapter cites specific uses of autoradiography in metallurgy and in biology. The former include studies of intergranular diffusion, heterogeneities of solidification, and the solidification of large ingots of metal (Fig. 1); localization of certain elements in a structure; and identification of precipitates and of the origin of inclusions. The latter include the determination of the conditions of fixation of a particular element; the study of cyclic absorption and elimination of certain compounds during the existences of living organisms; and the elucidation of the role of nucleic acids in the synthesis of living matter. (HPR)

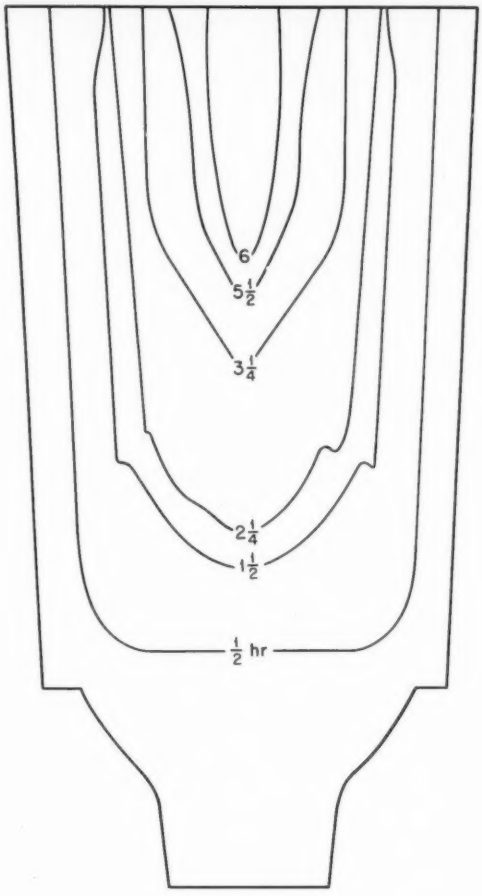


Fig. 1 Diagram of autoradiograph showing progress of solidification in a 30-ton steel ingot.

TEXTILE INDUSTRY

Bureau Eurisotop has probably published more material on the use of isotopes and radiation in the textile industry than in any other area. Two well-indexed bibliographies totaling more than 200 references, a monograph, bibliographic working documents consisting of nearly 1000 references, a series of four working documents comprising a total of nearly 200 individual reports and publications, and two proceedings of meetings make up the list to date.

Isotope Techniques and the Textile Industry, Parts I and II (in German), O. van Wauwe and D. Brandenburg, Eurisotop Booklet No. 6, Bibliographic Series No. 2, 201 pp., July 1965; Eurisotop Booklet No. 19, Bibliographic Series No. 5, 205 pp., May 1967.

The first of these compendiums contains 116 references and the second, 88. These are indexed under seven subject headings (Review Articles, Radioactive Tracers, Activation Analysis, Irradiation, Radiation–

Chemical Processes, Measurements by Radiation Absorption, and Static Elimination) and by ~1200 key terms. There is also an author index.

The Use of Radioisotopes in the Textile Industry (in French), P. Rochas and L. Gavet, Eurisotop Booklet No. 9, Monograph Series No. 2, 71 pp., September 1966 (191 references).

This booklet starts out with a general consideration of atomic structure, the method of detection of radiation and measurement of its intensity, and a listing of the various techniques applicable to the textile industry. Then specific examples of industrial and research applications to textile problems are delineated. These include fiber studies; operations involving textile transformations (e.g., spinning, weaving, oiling, sizing); handling and finishing operations (e.g., washing, dyeing, sizing); radiochemical analyses (e.g., determination of the function of free acids in cellulose, determination of the function of carbonyls in cellulose, determination of rate of hydration of cellulose); modification of textile structure and formation of textile copolymers by irradiation; and synthesis of textile graft copolymers by irradiation. The last topics consider a number of grafting agents, including styrene, acrylonitrile, methylmethacrylate, vinyl pyridine, acrylic acid, and maleic acid.

Irradiation Techniques in the Textile Industry, Collection of Abstracts, Vols. I and II (in English), L. Wiesnen (Ed.), Eurisotop Working Documents Nos. 52 and 123, ~500 pp., 1966 and 1970 (~1000 abstracts).

These compilations list the abstracts under the following headings: (1) Radiation Chemistry and Technology of Polymerization and Polymers; (2) Cellulosic Materials; (3) Wool and Other Noncellulosic Natural Fiber Materials; (4) Polyolefinic Materials; (5) Polyvinylic Materials; (6) Polyacrylic Materials; (7) Polyesteric and Polyoxymethylenic Materials; (8) Polyamidic Materials; (9) Polyhalogenated Materials; (10) Natural Rubber and Synthetic Elastomers; (11) Irradiation Effects by Electrons, Ions, and Neutrons. Also included are a Report Index, showing availability of the reports abstracted, and an Author Index.

Automation and Simplification in the Textile Industry Through Nuclear Radiation (in French, German, Dutch, or Italian), H. Maschner (Comp.), Proceedings of a Meeting, Evian-les-Bains, France, May 8–10, 1967, Eurisotop Booklet No. 35, Proceedings Series No. 10, 373 pp., 1967 (27 references).

The referenced meeting dealt primarily with radiometric problems associated with the use of nuclear techniques in the textile industry: (1) Measurement of Uniformity and Dimensions (11 papers); (2) Other Applications of Radiometric Methods (2 papers); (3) Moisture Determination (4 papers); (4) Control of Finishing Processes (5 papers); and (5) Density and Level Measurements (2 papers). One session was concerned with the Elimination of Electrostatic Charges (3 papers).

Radioisotopes and Radiation for the Improvement of Processing Techniques in the Textile Industry (in German), H. Maschner (Comp.), Proceedings of a Meeting, Baden-Baden, Germany, July 3–5, 1967, Eurisotop Booklet No. 36, Proceedings Series No. 11, 400 pp., 1967 (127 references).

The second conference on isotopes and radiation in the textile industry emphasized the use of radiochemical techniques in evaluating production processes, as well as in analyzing starting materials and end products, and the use of radiation in improving textile products. Sessions were held on (1) Investigations of the Distribution of Coatings (6 papers); (2) Investigations of Stainability and Removal of Stain (2 papers); (3) Investigations with Labeled Cellulose (4 papers); (4) Studies of Industrial Effluents (2 papers); (5) Analytical Methods (e.g., activation analysis of elements in textiles; continuous determination of zinc in spinning baths) (4 papers). The session on Radiation–Chemical Investigations included 6 papers (e.g., improvement in textiles; sterilization and disinfection; cross-linking of synthetic latex).

Textiles, Volumes A–D, Eurisotop Working Documents Nos. 1, 3, 5, 7 (plus Indexes 2 and 4), 1967.

In a 4-volume set, Bureau Eurisotop has put together 195 reprints of articles dealing with the various facets of the use of radioisotopes and radiation in the textile industry. The papers cover every conceivable area of application. Although the organization of the papers themselves does not seem to follow any particular pattern so far as content is concerned, the indexes are well detailed, making reference to any particular subject relatively simple. Author indexes are also included. The papers are in their original languages. (PSB)

WOOD–PLASTIC COMPOSITES

Bureau Eurisotop has prepared four booklets in their Monograph Series relating to the production of

wood–plastic composites (WPC). Background information on the anatomic structure, chemical and physical properties, impregnability, and effects of radiation on the wood itself is presented in addition to the description of the process—woods and plastics used, impregnation, and irradiation polymerization of monomers—and properties, fabrication, evaluation, uses, and economics of the product.

Impregnation of Wood (in German or French), A. Villière, Eurisotop Booklet No. 21, Monograph Series No. 5, 43 pp., November 1968 (18 references).

This booklet is devoted both to the general study of wood, which lends itself to the impregnation of synthetic materials, and to the methods of impregnation. The anatomic structure of wood is described in some detail, and attention is given to the two basic types of woods—one type from the conifers (resinous) which have a relatively simple structure and the other type from the leafy (foliar) trees which have a distinctly more complicated structure. Both the anatomic and the cellular structures strongly influence the impregnability of the wood. Three important interrelated physical characteristics, density, moisture, and dimensional stability (shrinking), must also be considered; hence the moisture–shrinking interaction is explained. The general process of impregnation of wet and dry woods with aqueous solutions and organic solvents and the industrial process for impregnation of railroad ties with creosote are described in some detail.

Because of the problems associated with the production of massive pieces of WPC (obtaining large pieces of choice, well-seasoned wood; necessity of using a vacuum in pressure impregnation; achieving homogeneous impregnation; etc.), the use of WPC veneer slabs appears to offer several technical and economic advantages: quality woods free of knots and irregularities are more easily obtained, drying of the veneer slabs to the desired moisture content is easy, uniform impregnation can be accomplished by a simple soaking procedure, etc. The assembly of multiple impregnated veneer-type slabs presents the possibility of fabrication of an extremely tough material; however, the monomer must serve to glue the plies together, and a method for irradiation of the slabs while being pressed together must be worked out. Persons beginning investigative studies are advised to work with the more easily impregnated woods, e.g., beech and birch. In the production of WPC, there are technological problems beyond those of the irradiation process itself.

Improvement of Wood by Monomer Impregnation and Radiation Polymerization (in German or French), A. Burmester, Eurisotop Booklet No. 22, Monograph Series No. 6, 47 pp., March 1969 (71 references).

This booklet aims to inform interested persons of the actual state of development of the technique for producing WPC so that by knowing the existing possibilities they can more easily make the decisions that are necessary for its utilization.

The chemical, physical, and mechanical properties, including a discussion of the absorption and desorption of moisture and its effects on wood, are given, as is a somewhat detailed discussion of both the fine structure and the impregnability of wood. This is followed by presentation of some of the fundamentals of radical polymerization of monomers and the nature of both gamma and electron radiations.

The literature on the effects of radiation on wood indicates that even small doses of radiation may have some degradative effect on the chemical components of wood, and this effect varies with the wood species. The chemical changes in wood caused by irradiation are reflected in the physical properties of wood, especially its moisture absorption. Within humidity limits of 25 to 92%, the moisture equilibrium of irradiated wood is less than that of nonirradiated wood; above 92% the irradiated wood absorbs more moisture. The mechanical properties of wood also are affected by irradiation as shown by numerous references in the literature. In general, depending on species, some mechanical properties are enhanced at lower doses of radiation; at increasing doses, degradation becomes predominant.

In the study of the changes in sensitivity of irradiated wood to attack by living organisms, the general tendency of higher doses of radiation to decrease the resistance of wood to termites and fungi was found to vary with the wood species and the organism.

Before going into the study of wood–plastics, a discussion is presented on the interaction of radiation on cellulosic materials and monomers. Irradiation of the cellulose prior to contact with the monomer (rather than simultaneous irradiation of cellulose and monomer) appears to improve the grafting process. The relationship of water and other swelling agents with cellulose is also considered.

Wood–plastic composites prepared from several wood species and various monomers are reviewed. This review includes discussions of WPC made from several acrylates, acrylonitrile, diisocyanate, formaldehyde, styrene, vinyl acetate, and vinyl propionate as well as

a consideration of physical properties, mechanical properties, additives, and remaining research on process techniques.

A New Material: Wood–Plastics (in German or French), F. Rodenay, Eurisotop Booklet No. 23, Monograph Series No. 6, 14 pp., November 1968 (16 references).

The third booklet, No. 23, describes wood–plastics as a new material and the work done by the Société Conservatome Industrie, which has been limited to the use of methylmethacrylate (MMA). The procedure used is rather simple: the wood is first degassed under vacuum to facilitate the penetration of monomer, and MMA (which has a viscosity about that of water) is then introduced, covering the wood and penetrating easily into it. After impregnation, the excess monomer is removed and the impregnated wood is kept under a nitrogen atmosphere; a certain delay before irradiation favors the diffusion of monomer into the wood. The rate of reaction can be controlled by the dose rate. Irregularities caused by surface depletion of monomer can be removed by sanding; for this reason, shaped pieces are impregnated and the monomer polymerized before final finishing is done.

The most notable changes are the improved appearance and dimensional stability (low rate of moisture absorption) and the enhanced mechanical properties. In brief, the new material retains the richness of wood yet has the resistance of plastics. Ordinary woodworking tools can be used for WPC; however, tools used for working decorative laminates, improved or reconstituted woods, or plastics are recommended. Wood–plastics cannot be nailed (without predrilling) because of splitting; they may, however, be glued.

The market for wood–plastics has not developed; to be economic, it must be produced on a large industrial scale, which may reduce the cost considerably. The price of wood–plastics is comparable to that of the most expensive of the exotic woods. In a detailed economic study, one must consider other factors:

- Reduction of the drying time and thus of the wood inventory.
- Education of the clientele to the advantages of WPC with respect to reduced maintenance and the possibility of prefinishing product items.
- Industrialization of some manufacturing that is now done by handcrafters.
- Possibility of treating pieces that have already been roughly shaped to conserve monomer.
- Simplified finishing processes: requires only sanding and polishing.
- Possibility of having the treatment subcontracted, thus reducing the investment cost for numerous small enterprises in the wood industry.

In France in 1966 a potential market survey of WPC was made in the wood industry. The results of this evaluation are presented in the booklet—in general, wood–plastics were an improvement over wood with their appearance, maintenance, stain and scratch resistance, dimensional stability, weather and water resistance, etc; however, their weight, inability to be nailed, and, in some cases, price, were unfavorable factors.

Mosaic parquet flooring is a particularly interesting potential application because of the many desirable properties of wood–plastic and the ease of maintenance. For this reason Euratom has considered the development of wood–plastics in this application.

Impregnated and Irradiated Wood Plastics (in German or French), A. Villière, Eurisotop Booklet No. 24, Monograph Series No. 10, 35 pp., January 1969.

This booklet, a supplement to Booklet 22, gives the results of tests on the physical properties of WPC conducted by the Technical Wood Center in agreement with the Commissariat à l'Énergie Atomique and in collaboration with the Société Conservatome Industrie; it is intended to help introduce the new combination—wood–plastic—obtained by irradiation. The goal of the testing was to determine, on the one hand, the physical and mechanical properties of these new materials and, on the other, to study their capabilities with respect to gluing, finishing, behavior in fire, and resistance to biological agents.

In these studies of WPC, three wood species—pine, beech, and oak—were used and methylmethacrylate was used as the impregnating monomer. For the mechanical tests—compression, flexure (bending), impact, etc.—standard French test pieces, previously sawed to the needed dimensions, were used, and, for all other tests, 10- by 10- or 20- by 20-cm plaques 1 cm thick were prepared. In addition, some beech veneers 20 by 20 cm by 2 mm thick and some gaboon laminates of 10 mm thickness were also tested. Before impregnation, samples were stabilized and weighed in an atmosphere of 20°C and 60% relative humidity, and, after impregnation and irradiation, samples were again stabilized and reweighed. The monomer absorbed and irradiated was thus accounted for. An equivalent

number of controls were retained for all samples. Impregnation was done by the Société Conservatome Industrie. For beech, only the vacuum method was used, but, for oak and pine, the vacuum–pressure method was used. The IRMA irradiator of the Société Conservatome Industrie, Saclay, was used to give the samples a dose of about 1.5 Mrads in 5 to 15 hr.

The amount of monomer absorbed by the wood by impregnation was on the average 30 to 40% for oak, 70 to 75% for beech (blocks), 56% for beech veneers, and 95% for pine (based on the initial weight of wood stabilized at 20°C and 65% relative humidity).

Various physical characteristics of wood—density, moisture equilibrium, shrinking, etc.—were determined on plaques 1 cm thick for three species—oak, beech, and pine—and on beech veneer 2 mm thick.

The observed density of WPC samples was slightly less than that calculated from the weight of monomer absorbed; this was the result of a slight swelling of the wood by the monomer. The length of the impregnated specimen appears to have no significant effect on the density.

There is considerable discussion of the phenomenon of shrinking (dimensional stability). In the stability test, the three woods were used—both treated (WPC) and untreated—and tests were made by exposure to atmospheres of controlled temperature and humidity and by immersion in water. In general, WPC had about 50% less dimensional change than that of untreated wood. In similar tests on beech veneers, the impregnated material shrank 47% less than that of the same untreated veneer.

In the water immersion test, where only the beech veneer was used, the wood–plastic veneer swelled 44% less than the untreated controls. On a weight basis, if one corrects the weight of the WPC to only the weight of wood (not the weight of wood plus plastic), the controls absorbed 110% of their weight and the WPC 50%. These tests are described in detail in the booklet, which also gives some discussion on equilibrium moisture content of both treated (WPC) and untreated woods.

The mechanical tests were made on the untreated controls and on WPC according to the procedures of the French AFNOR, which uses test pieces of standard dimensions.

Before testing, all samples were conditioned in an atmosphere of 20°C and 65% humidity. The moisture equilibrium was 12%. The outstanding increases in mechanical properties are shown as improvement over natural untreated wood as follows:

Test	Beech	Oak
Compression	110%	54%
Static flexure	67%	35%
Modulus of elasticity	39%	18%
Hardness	330%	220%

In a complementary test, screwing and nailing of WPC was studied. Screws could only be driven in pilot holes of the correct size (too small holes caused breakage of the screws). The holding power of screws in WPC is significantly greater than that for natural wood. The test showed that WPC could not be nailed.

In a test with fungi on beech and pine WPC, the beech showed little attack; however, impregnation of pine with plastic did not afford protection against fungal attack.

Resistance to attack by termites was also tested. Results were particularly satisfying for oak and a little less for beech (but still very good), and the resistance of these two WPC woods rivaled that of some of the tropical woods which are invulnerable to the action of insects.

In fire tests, the flammability of WPC is slightly retarded; however, when WPC is ignited, it burns longer and therefore in essence offers no fire protection.

In gluing tests on oak WPC, vinyl emulsion and poly(butyl acrylate) glues were not satisfactory since samples under test ruptured in the plane of the glue. For beech, an improved urea glue and a resorcinol glue were used, and the resorcinol glue gave relatively suitable results. In gluing oak WPC to chipboard, the classic vinyl glues gave satisfactory results.

Finishing of WPC can be accomplished by simple sanding, buffing, and polishing. However, in exterior applications, the surface undergoes some weathering. In interior applications (furniture, etc.), a high surface lustre can be achieved by polishing without varnishing or waxing. In a test on staining of the finish, the plastic-impregnated woods did not show as much resistance as woods finished with cellulosic and polyurethane varnishes. In WPC the wood is impregnated in the interstices and the wood membranes remain uncovered and unprotected; this permits substances to penetrate easily as in a natural wood, thus causing stain. On the other hand, the cellulosic and polyurethane finishes form a continuous coat that prohibits the staining material from entering the wood structure. However, although their protection is not as good, the aesthetic surfaces obtained with WPC are comparable to those produced with modern finishing agents.

(REG)

FOOD INDUSTRY

Control Method for Research on Salmonella in Forage Decontaminated by Irradiation (in German, French, Italian, or Dutch), E. H. Kampelmacher and D.A.A. Mossel, Eurisotop Booklet No. 20 (2nd edition), 7 pp., 1969.

This pamphlet is a compilation of standard methods of microbiological analysis agreed upon at a work session of members of the European Community Commission (organized by the Bureau Eurisotop) in Brussels, Nov. 1, 1967. The methods are outlined in four sections: research on salmonella in mixed forage; counting of enterobacteriaceae in unirradiated forage; detection of enterobacteriaceae in irradiated forage; and culture mediums to be used. Use of the methods will allow the different laboratories in the Community to obtain comparable results. (FEM)

The Irradiation of Potatoes (individual papers in French, German, or English), Proceedings of a Symposium, Brussels, Oct. 1–2, 1969, Eurisotop Booklet No. 45, Proceedings Series No. 18, 165 pp., 1970.

At this symposium, researchers from Belgium, West Germany, France, Italy, the Netherlands, and the European Community Commission (ECC) gathered to review the progress made in studies on the irradiation of potatoes.

The economic importance of the production, use, storage, and loss of potatoes to the European countries was reviewed by Besnard (ECC), who pointed out that production in 1967–1968 was about 41×10^6 metric tons, with West Germany growing nearly 48% of the crop. Loss currently amounts to about 5%.

The second session of the symposium was devoted to the scientific, technical, and wholesomeness aspects of the sprout inhibition treatment. Patzold (West Germany), in a paper with 97 references, reviewed the advantages and disadvantages of the different sprout inhibition methods, comparing each method to irradiation. Heilinger (West Germany) presented data from experiments using low doses (less than 3200 rads) at high intensity on both sprouted and unsprouted tubers. Special attention was paid to carbohydrate metabolism. Respiration and weight loss were covered in a paper by Nys (Belgium), who said that, while a dose of 8 krads is theoretically sufficient, dosimetry and dose distribution inequities require a dose of 10 to 12 krads. He also pointed out that chemical treatment is still less expensive than radiation. Experiments using ^{60}Co and ^{137}Cs showed that potatoes sharply increase respiration immediately after irradiation; there is also an increase in soluble sugars, according to Sandret (France). Irradiation forms substances that diffuse rapidly and inhibit growth; parenchyma-cell cultures were inhibited by an extract from irradiated tubers. Stahl (West Germany) reviewed fogging methods that control not only sprouting but also development of rot to some degree. The results of a consumer survey were presented by Diehl and Grunewald (West Germany). Hansa variety potatoes were irradiated with a 10-MeV accelerator to a dose of 11.7 krads and then distributed to families along with a detailed questionnaire. Replies were assigned numerical values and plotted; the graphs show that preference for irradiated potatoes increased with storage time because controls, of course, became lower in quality as they sprouted. Results were characterized as "very positive."

Two papers were presented in a session on economic and commercial aspects of potato irradiation. Vidal (France) pointed out that permission had been granted to irradiate potatoes commercially in Canada, USSR, USA, and Israel. He stated that a cost increase of 5 to 12% would be considered acceptable. Economic factors include whether or not consumption per capita is increasing or decreasing and who bears the cost of treatment. Vidal also stated that users of large quantities, e.g., processing plants, would be more willing to pay costs of keeping supplies in top quality than would an individual consumer. Consumers have reacted favorably both to the actual product and to a survey in Greenland, Canada, and Israel. Ulmann (Netherlands) presented results of a recent marketing of irradiated potatoes. Dutch production is about 5×10^6 metric tons/year, of which about 27% is exported. The advantages of large-size plants were pointed out; but beyond a certain point, large size starts creating its own problems. Ulmann also pointed out the special label symbols used in Israel and the Netherlands for irradiated food.

There are three summaries of panel discussions; these covered scientific and technical questions, commercial and economic questions, and regulatory and wholesomeness questions. (FEM)

The Application of Irradiation Techniques in the Food Industry (in French), R. Mulkens (Ed.), Proceedings of a Conference, Brussels, Feb. 23–24, 1967, Eurisotop Booklet No. 43, Proceedings Series No. 16, 239 pp., 1971.

This conference, sponsored by Eurisotop and attended by 85 scientists of the European Community, completely reviewed the situation in regard to the application of radiation for preservation of foods.

There were 12 sessions: (1) general exploration of the problem and status review of various countries, particularly those of the Community; (2) application of irradiation techniques to bulbs, roots, and tubers; (3) application of irradiation techniques to cereals and cereal products; (4) application of irradiation techniques for pasteurization of fruits and vegetables; (5) application of irradiation techniques to dehydrated vegetables; (6) application of irradiation techniques for the treatment of wines; (7) application of irradiation techniques to fresh meats; (8) application of irradiation techniques to seafoods, seafood products, and canned meats; (9) irradiation technology; (10) wholesomeness and legal aspects of irradiation; (11) industrial initiatives in member states of the Community; and (12) suggestions for action to be taken by member states.

In each session, there were prepared papers, but an important point is that the majority of the booklet is devoted to recording the floor discussions, which were quite lively, on each aspect of the subject. The general conclusion reached was that the problems remaining before commercialization and full-scale use of the technique are more administrative than they are scientific or technical. (FEM)

HYDROLOGY

Hydrological Applications of Radioactive Isotopes. Federal Republic of Germany. Status up to 1965 (in German, French, or Italian), H. Moser, F. Neumaier, and W. Rauert, Eurisotop Booklet No. 16, Bibliographic Series No. 3, 21 pp., 1966.

This booklet is a concise state-of-the-art review up to 1965 on the uses of radioisotopes in hydrology in the Federal Republic of Germany. Subjects covered are the age determination of groundwater using the ^{14}C method, hydrological tritium investigations, measurement of the flow rate and behavior of rivers, investigation of sand migration in rivers and coastal waters, tracer methods for groundwater, and application of gamma radiation and neutrons for determination of soil density and moisture. The review is supported with 81 references. (REG)

Portable X-Ray Spectrometer: Applications to Field Geology in the Galápagos Islands

By C. A. Ziegler,* T. Simkin,† and J. M. Brinkerhoff*

Supplementary Keywords: *geophysics; X-ray fluorescence; spectrometry; source, X ray; method; comparison; theory, applied; data handling; Ca; Fe; K; Ti.*

Abstract: *Portable X-ray spectrometers have been used widely in ore prospecting and processing where repetitive measurement of a single heavy element is required. However, they have been used little by research geologists, who generally are interested in multielement analysis for the lighter elements. The performance in the field of a portable X-ray spectrometer that can measure light elements (atomic weights down to 14) is described, together with a new operational format that provides a simplified method of multielement analysis. The spectrometer and associated equipment are sufficiently lightweight (21 lb) to be readily backpacked to remote sites. They allow samples to be rapidly assayed in the field (four elements per sample can be measured in 20 min). The applications described for the spectrometer in geological research—specifically in volcanology—illustrate the value to the field geologist of obtaining immediate, on-site, quantitative geochemical information. The X-ray spectrometric data agree well with wet-chemical data.*

Modern geology increasingly emphasizes laboratory work—a situation that has had important effects on field work. Because laboratory analysis has revealed significant differences among specimens not easily distinguished in the field, the field geologist now has greater difficulty convincing himself that he has collected a truly representative suite of specimens. At the same time, the increasingly sophisticated analyses made on his specimens emphasize the importance of wise choice by the field collector. With only classical field equipment, significant differences among speci-

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mens will often go unrecognized until the specimens are in the laboratory; some differences will never be recognized because the critical specimen was not collected. Ideally, repeated visits between laboratory and field eventually yield a fine collection of specimens, but this procedure is obviously difficult when the field is remote.

In this study, active volcanoes in the Galápagos Islands were the remote and difficult site for field-testing a portable X-ray spectrometer from June 16 to 30, 1971. Quickly chilled volcanic products are commonly too fine-grained for effective megascopic classification, and the volcanoes under study (uninhabited Pacific islands 1000 km west of Ecuador) present logistic difficulties that encourage maximum use of field time. Classical techniques were used to study the recent caldera collapse of Volcan Fernandina and the volcanic ash deposits of Fernandina and Marchena. But the instrumental innovation of this expedition was the use of a portable X-ray spectrometer that permitted field accumulation of geochemical data.

BACKGROUND

In geochemistry, the X-ray fluorescence spectrometer has long been a laboratory tool to measure elemental composition. Hence it is not surprising that, when small portable versions of this instrument (based on the use of radioisotope-excited X rays) became commercially available in 1964, measurements of geological samples were among its earliest uses.[1-3] These applications involved mineral prospecting, since portability allowed the extension of X-ray spectroscopy to on-site detection and assay of ores of most of the common metals. In the intervening years, this instrument has been used further in prospecting and mining technology. Its chief advantage to the mineral prospector is its ability to immediately provide assay data, which can be used to optimally direct sampling operations. This capability should be equally advantageous to the research geologist. However, aside from ore prospecting, few geological applications have been reported. Indeed, the only measurements of common terrestrial rocks, made with this instrument, were undertaken to ascertain its applicability for in situ measurement of lunar rocks.[4]

There are important technical reasons why this instrument has not become a general-purpose tool for the field geologist:

1. Prospecting and assay involve measurement of a single heavy element in a matrix of lighter elements, e.g., tin in its ore. However, in geological research, there is need to measure several light elements (atomic weight <20) in an admixture of light and intermediate-weight elements. Until recently, portable spectrometers depended on scintillation counters to detect X rays. Such detectors cannot detect the low-energy X rays from elements of atomic weight <16, and their performance is often inadequate for elements of atomic weight <20.

2. Operational techniques evolved for prospecting and assay require instrument calibration with a set of ore samples, previously analyzed by wet chemistry, to construct a calibration curve, which applies only to a single, sought element. Additional curves are needed to display the instrument response to this element in each matrix type expected to be encountered. Although this approach is adequate for repetitive assay for a single element (as in prospecting), it is cumbersome for the research geologist, who generally wishes to measure several elements in various matrices.

The first objection was eliminated by the recent (1969) commercial availability of a portable X-ray spectrometer that can measure light elements. This instrument uses a ruggedized thin-window proportional counter, which can measure fluorescence X rays from elements of atomic weight down to 14.* The second objection can be overcome by the development of a simple operational format and of methods for calibrating the portable X-ray spectrometer that are specially designed to allow multielement field analysis of geological samples.

This article presents such a new operational format together with data that demonstrate its implementation in geological field research.

SITE ENVIRONMENT AND ITS EFFECT ON EQUIPMENT REQUIREMENTS

The equatorial climate of the Galápagos Islands is moderated by locally cool ocean temperatures due to the Humboldt current; the recorded local air temperatures during periods of actual operation of the X-ray spectrometer varied from 78 to 86°F. The islands studied (Fernandina and Marchena) are uninhabited and are accessible only by means of small boats that can land directly on the beach. These difficulties cause expeditions that last less than several weeks to be

*The Panalyzer 4000 X-Ray Spectrochemical Analyzer made by Panametrics, Inc., 221 Crescent St., Waltham, Mass. 02154.

uneconomical. Thus, the equipment must be usable for weeks without requiring off-island supplies. Because bulky apparatus and fuel are difficult to land, use of a general-purpose generator of electricity is undesirable; hence, electrical instruments required a self-contained power source. The extremely rugged terrain precluded the use of wheeled vehicles; therefore, all equipment also had to be suitable for backpacking over reasonable distances.

EQUIPMENT

Numerous published papers, including a review, describe in detail portable spectrometers based on radioisotope-excited X-ray fluorescence.[5] Therefore, the theory of operation of this instrumentation need not be discussed. In the work reported here, the Panalyzer 4000 was used. It can be operated from a-c power mains (used during calibration in the laboratory) or from an internal battery pack (used to make field measurements). The battery pack uses 10 ordinary D-size flashlight batteries, which can power the unit for a week of normal usage. The instrument is supplied with two carrying cases—one for the electronic console and its internal battery pack (15½ lb) and a second that contains the measuring head and accessory equipment (sample holders, filters, and cables), which weigh 6 lb. Both cases are supplied with straps for short portages, and both can be stowed easily into a single standard backpack for transport over longer distances. Figure 1 shows the complete unit being deployed from a backpack. Additional equipment for sample preparation consists of a hammer, a percussion mortar and pestle, and a sieve.

Fig. 1 Field deployment of the portable X-ray spectrometer (Panalyzer 4000). (Note that the entire unit can be fitted into a single backpack and that all components are stowed in two cases provided with their own carrying straps for short portages. Site is northwest sector of Fernandina.)

ELEMENTS MEASURED

The lava that produces the rocks and ash deposits of these islands is dominantly basalt. The minerals that crystallize from it are largely silicates, and the elements present are, in order of decreasing abundance: O, Si, Al, Fe, Ca, Mg, Na, Ti, K, P, and Mn. The four elements selected for field determination were Fe, Ti, Ca, and K; Mn and P are present in very small concentration; Si shows less relative variation in basalts than do the four elements chosen; and the remaining elements are too light for analysis with the Panalyzer 4000. (An improved low-energy detector, now being developed, should make determinations of Al, Mg, and Na possible in the near future with a modified version of this instrument.) Previous reconnaissance studies of the Galápagos Islands[6] indicated that total Fe content (as FeO) ranges from 4 to 14 wt.%, with that of most of the ash (and common rocks) falling between 11 and 12%. The Ca content (as CaO) also shows a broad range (1 to 12%) but is much influenced by the presence of early formed feldspar crystals and, like Fe, is more useful in indicating unusual rock types than in distinguishing significant differences among common rocks. On the other hand, Ti and K, although much lower in total abundance, vary considerably in amount within the common rocks; these variations are significant in terms of rock origins. In common Galápagos basalts, reported contents range from 1.5 to 3.0% TiO_2 and from 0.2 to 1.0% K_2O; most K values were below the limit of detection (0.2%). After the four elements had been selected for measurement, the spectrometer was calibrated and was supplied with X-ray filters and with standards prior to the expedition's departure.

OPERATIONAL FORMAT FOR FIELD USE

A simple format for data reduction in the field was developed and used in this survey. The operational system involves using calibration and setup procedures performed in the laboratory to evolve software, which eliminates the need for field interpolation involving a number of calibration curves. This, in itself, is a significant practical advantage for multielement analysis. However, this method involves more than merely substituting analytical for graphical procedures. Other desirable features are:

1. "Wide-range calibration." Conventional graphical methods require previously prepared calibration curves. Hence, if samples that fall outside the expected range are encountered in the field, the curves are useless. The analytic format presented here allows the sought element to be measured over a very wide concentration range. Thus, accurate knowledge of the variation of the sought element in the field samples is not required—an important advantage, for example, in petrological reconnaissance, where such variations may not be predictable with precision.

2. "Automatic correction for variations in matrix radiation." For graphical procedures, the intercept of the calibration curve is assumed to remain constant. This condition holds only if matrix radiation flux from all measured samples remains the same. No such assumption is made in this operational format. Rather, the radiation from the principal matrix materials, e.g., alumina and silica, is measured specifically for each sample in a separate energy channel. The format uses this information to correct for variations in matrix radiation.

3. "Need for only one calibration sample."* Graphical methods require at least two (and preferably three to five) calibration samples that span the expected concentration range of the sought element. The operational procedures presented here allow calibration over a very wide concentration range by use of only *one* calibration sample that contains a typical concentration of the sought element. This feature significantly reduces the amount of independent laboratory analysis needed for calibration and is an important factor for geochemical applications involving multielement assay.

The laboratory procedures for calibration and preparation of the data required in the field are described in detail in the appendix. From these data, software was developed specifically for field use (Fig. 2). The aim was to provide a record form that contains not only spaces for inserting field data but operational symbols and system constants that, when applied to recorded data, lead directly to a numerical value for weight percent of the sought element. Also, the procedures required to fill out the information on the top part of the form involve measurement of an elemental standard and a scattering standard (Teflon) and require the operator to make comparisons with calibration data. This feature prevents usual operator errors, e.g., forgetting to change the X-ray filter.

Figure 2(*a*) shows the basic form. This form is modified by insertion of system constants derived from laboratory calibration as described in the appendix. Figure 2(*b*) shows a sample of the form, with system

*The term "calibration sample" refers to a sample, typical of those to be encountered in the field, in which the concentration of the sought element has been determined accurately, as, for example, by wet chemistry.

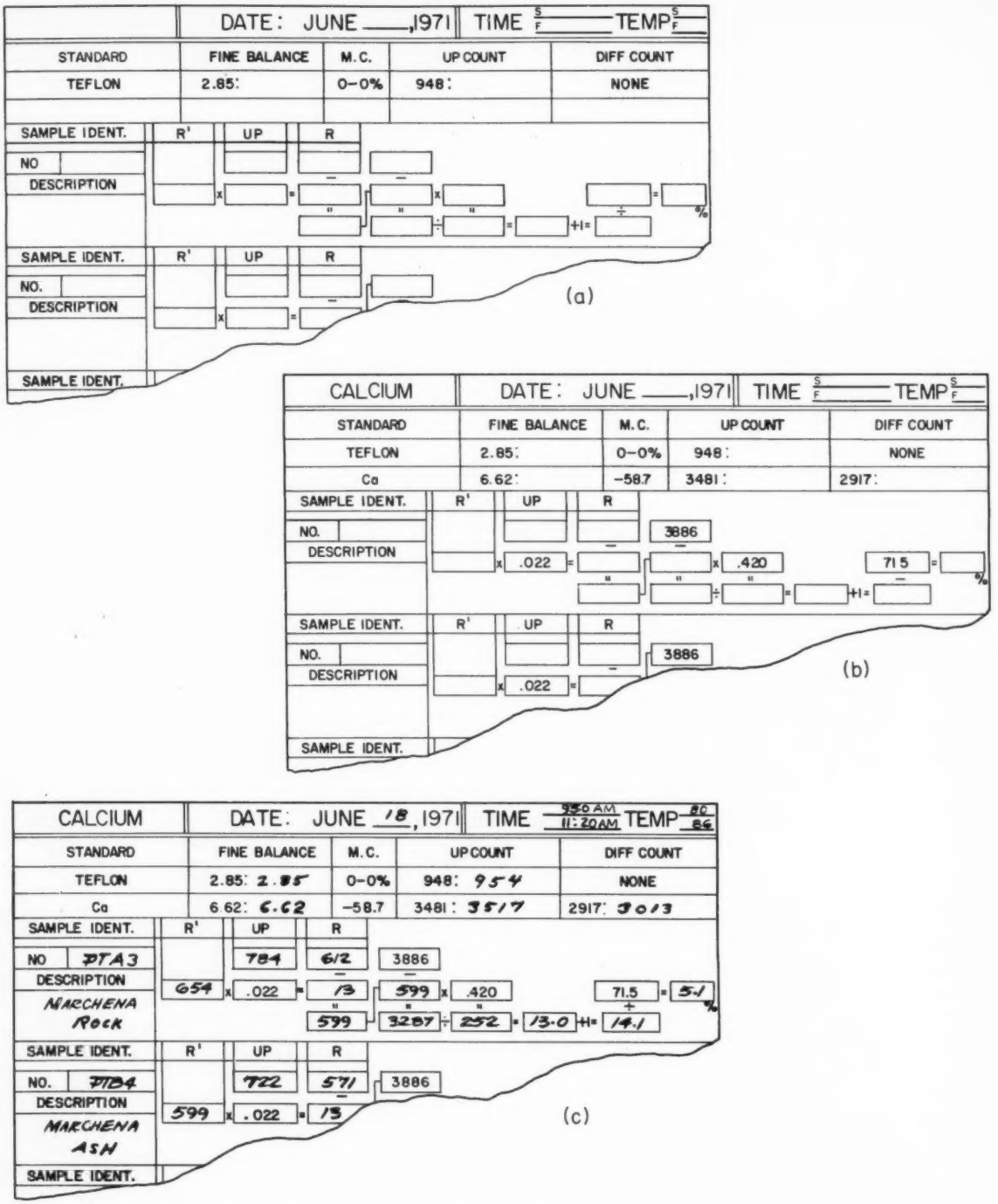

Fig. 2 Software developed for field analysis of geological samples: (*a*) the basic form, (*b*) the form after the insertion of system constants derived from laboratory calibration—this is the version carried into the field, and (*c*) the final form as it appears after execution. (Note that the *UP* count is not used in the data-processing format but is needed later to calculate the measurement uncertainty.)

constants for calcium. Three similar forms with appropriate constants for K, Ti, and Fe were also prepared. These forms, which contained laboratory data, were then reproduced to provide a suitable number of blanks for field measurements. In use, the operator pencils in the observed numerical values of R', UP, and R. After measuring a given number of samples, the operator simply performs the indicated arithmetic to obtain the weight percent, as shown in Fig. 2(*c*). The form is structured to eliminate the need to use scratch paper for additions and subtractions. A slide rule is used for multiplications and divisions. A single 8½- × 11-in. page that contains blocks for measuring seven samples was used in this survey since it was found convenient to perform sample measurements in batches of seven, corresponding to a total measuring period of 1 hr. For each of the four measured elements, three 16-sec counts are taken for each sample (corresponding to R', UP, and R), but the total measuring period of 1 hr for seven samples includes activities such as changing filters and recording data. An additional 80 min is required to perform (and double check) the arithmetic for one batch. Therefore, on the average, a total of 20 min per sample is required to arrive at the weight percent of each of the four sought elements (8½ min for measuring; 11½ min for calculation).

SAMPLE PREPARATION IN THE FIELD

All samples, including rock samples, were measured in powdered form. The importance of particle-size distribution on elemental assays by X-ray fluorescence analysis is well known.[7] In this work, it was established empirically that reproducible results can be obtained by crushing all samples until they passed through U. S. Standard Sieve Mesh No. 325. This ensured that all particles were $<44\ \mu$ in size. Sample-preparation times (by use of a percussion mortar and pestle and No. 325 sieve) varied from a few minutes for ash samples to 20 min for the harder rocks.

TYPICAL FIELD RESULTS

During this survey a total of 37 samples were prepared and measured in the field; however, only those typical of specific applications are presented here.

Sampling Technique

The first field use of this instrument was to test the chemical homogeneity of the ash deposits under study. Specimens from two contrasting locations of the 1968 Fernandina ash deposits were analyzed (both bulk samples and fractions having different sizes); Table 1 gives the results, along with comparable measurements from Isla Marchena.* The accuracy of these measurements is such that variations of ±0.2 wt.% can be taken as significant. These field results suggest that

Table 1 Geochemical Variation of Ash with Particle Size

Location and sample number	Particle size (N), μ	Element, wt.%			
		K	Ca	Ti	Fe
Fernandina No. 1	Homogeneous	<0.2	8.6	1.2	7.6
	44 < N < 88	<0.2	8.5	1.3	7.7
	350 < N < 710	<0.2	8.3	1.2	7.9
Fernandina No. 2	Homogeneous	<0.2	8.4	1.3	7.2
	350 < N < 710	<0.2	8.6	1.4	7.2
	N < 88	<0.2	8.2	1.2	7.4
Marchena No. 1	N < 62.5	0.4	7.6	1.2	9.1
	62.5 < N < 250	<0.2	7.1	0.9	8.2
	250 < N < 1000	<0.2	7.6	1.0	8.2

To demonstrate the simplicity of using this format, a learning trial was made with persons capable of using a slide rule but otherwise completely unfamiliar with the instrument and its theory of operation. Such persons measured samples and processed the data to the point of producing final assay results with <30 min of instruction (the assay data reported here were, in part, obtained by such persons).

*Particle-size distribution of ash was determined on-site by use of a set of 3-in.-diameter sieves and a portable weighing device. A field microscope was used for petrographic analysis.

Fernandina ash composition does not depend on the size-fraction analyses. This finding helped to modify the subsequent sampling of Fernandina ash. When fractions of different size show significant differences in composition, it is obvious that a larger quantity must be collected for later laboratory study than when the ash is essentially homogeneous. The field results indicated that the collection of smaller quantities of material would be satisfactory. Since there were limits on the size of the collection, this freedom allowed the collection of more (and smaller) ash samples than would otherwise have been taken. The result was a more complete sample coverage than would have been possible without the data of Table 1. In addition, these data confirm the basaltic composition of all three samples, demonstrate small composition differences between Fernandina and Marchena ash (see also Table 2), and suggest that the juvenile glass component of the Fernandina ash is compositionally equivalent to the older lithic fragments with which it is associated in the ash deposit (microscopic investigation shows that glass is concentrated in the fine size fractions, yet all size fractions are essentially of the same composition).

Table 2 Comparison of Ash Samples from Various Islands

Location and sample number	Description	Element, wt.%			
		K	Ca	Ti	Fe
Fernandina	Average of five samples	<0.2	8.4	1.2	7.6
Marchena	Average of four samples	<0.2	7.6	1.1	8.7
Pinta No. 1	Surface ash	0.5	7.7	1.5	8.5
Pinta No. 2	Native ash	0.2	7.6	1.1	7.6

Correlation of Ash Deposits

The prehistoric eruption that left such massive ash deposits on Marchena would be expected to have left some record on the neighboring island of Pinta, 40 km downwind from Marchena. After collecting (and analyzing) on Marchena, the expedition made a brief stop at Pinta, where two generations of ash deposit were collected. Although both samples are clearly basaltic (Table 2), neither shows good chemical correlation with Marchena ash. Had good correlation been shown, however, having such chemical data while still in the field (where more detailed investigation could have been undertaken) would have been a great advantage.

Rock Analyses

The X-ray spectrometer can analyze bulk rocks as well as ash samples; Table 3 gives data from rock analyses. Significant lava flows and distinctive lithologies could be analyzed in the field, where there was still time to learn from the data and to let this new information guide, when necessary, the continuing field work.

Table 3 Results of Analysis of Rock Samples*

Sample identification	Element, wt.%			
	K	Ca	Ti	Fe
1	<0.2	8.8	0.6	5.7
2	<0.2	7.9	0.9	8.6
3	<0.2	8.5	0.9	8.0
4	<0.2	8.6	0.8	7.9
5	<0.2	7.6	1.1	8.5
6	<0.2	7.9	0.9	8.1
7	<0.2	67.2	0.6	<0.2

*It will be noted that the first and last samples of this batch differ sufficiently in composition to warrant collection for further examination.

COMPARISON WITH OTHER ANALYTICAL METHODS

The "calibration sample" was measured by wet chemistry and by other conventional laboratory methods to accurately establish known values for the concentrations of the four sought elements. All reported X-ray-analysis data were obtained with this calibration. A comparison was desired among the results obtained with the portable X-ray spectrometer and those obtained with other analytical methods. Accordingly, four samples (in powdered form) were selected and split into two equal portions. These portions were sent to two qualified independent testing laboratories for analysis; Table 4 gives the results.

The maximum errors claimed by L-1 and L-2 for Ti, Ca, and Fe are ±0.15 and ±0.1 wt.%, respectively. That of the X-ray method is ±0.2 wt.%. For Ca, Ti, and Fe, the results of both laboratories and of the X-ray method agree within these stated errors. Laboratory L-1 used wet chemistry for Ca, Ti, and Fe, whereas L-2 used atomic absorption spectrophotometry. Both L-1 and L-2 used atomic-absorption spectrophotometry to measure K. The maximum error reported for the

Table 4 Comparison of X-Ray Results with Values Obtained with Other Analytical Methods

Sample No.	Element, wt.%											
	K			Ca			Ti			Fe		
	L-1*	L-2*	X†	L-1	L-2	X	L-1	L-2	X	L-1	L-2	X
1	0.023	0.08	<0.2	9.04	9.2	8.8	0.99	1.2	0.6	5.41	5.6	5.7
2	0.074	0.37	<0.2	8.30	8.3	8.3	1.38	1.3	1.1	8.25	8.1	7.8
3	0.048	0.37	0.3	8.32	8.5	8.6	0.98	1.3	1.2	7.98	7.9	7.9
4	0.52	0.70	0.5	7.67	8.0	7.7	1.48	1.5	1.5	8.97	8.8	8.5

*L-1 and L-2 indicate the two independent testing laboratories.
†X indicates values obtained with the Panalyzer 4000.

atomic-absorption measurement of K was ±0.03 at 0.5 wt.% and ±0.01 at 0.02 wt.%. The results reported by L-1 and L-2 for K do not agree within this stated limit. The sensitivity of the X-ray method is 0.2 wt.% K; hence results lower than this value are not reported numerically but simply indicated as "<0.2 wt.%." Table 4 indicates that meaningful measurements by both X-ray and atomic-absorption methods were obtained only when the concentration of K was of the order of 0.5 wt.% and that the uncertainty was about ±0.2 wt.%. It is interesting to note that the data of Table 4 are compatible with variations observed by others in a comparison of the various results reported by a number of analytical laboratories who were asked to analyze geological samples.[8]

SUMMARY

The utility of a portable X-ray spectrometer that can measure light elements has been demonstrated by making in situ measurements on two of the Galápagos Islands. A new operational format for use with the spectrometer was developed and demonstrated; it allows multielement analysis to be done readily in the field without the use of conventional instrument-calibration curves. The results agree well with those obtained by wet-chemical methods of analysis.

ACKNOWLEDGMENT

Participation in this expedition was under the auspices of Educational Expeditions, Inc., of Belmont, Mass., in cooperation with the Smithsonian Institution. Acknowledgment is gratefully made to R. Miller and Eleanor DeChadenedez for help in sample preparation and measurement and to A. Bress for the photography. (HPR)

APPENDIX

Wide-Range Calibration

Wide-range calibration of the system is based on the formula

$$R'' = \frac{Ap}{p + (1 - p)k} \tag{1}$$

where R'' = output due to element being measured
p = fractional weight-abundance of the element being measured
A = output for a 100%-abundant sample of the element
k = calibration constant

This formula follows from a simple model in which fluorescence-producing radiation is considered to enter the sample and to be absorbed exponentially. The X-ray fluorescence produced is then considered to be similarly absorbed on its return path to the radiation detector. From Eq. 1 it follows that

$$p = \frac{1}{1 + \left(\frac{A - R''}{kR''}\right)} \tag{2}$$

It may be further shown that the quantity A in Eqs. 1 and 2 may also refer to a *compound* of the element being measured, in which case the quantity p is replaced by the quantity p/C, where C is the fractional abundance of the given element in the compound. In this case, we obtain the more general relation

$$p = \frac{C}{1 + \left(\frac{A - R''}{kR''}\right)} \tag{3}$$

which is the formula on which the worksheet computation is based.

The particular compound chosen for use in the calibration need not actually be present in the matrix. It is sufficient merely that the other elements of the compound, or similar elements, be present in the matrix in at least the same abundance relative to the element being measured and that the remaining matrix elements be of the same general character. In practice, certain rather broad substitutions may be made, especially among the lighter elements (C, N, O, H, etc.), it being required only to preserve the same absorption coefficient per unit atomic weight. Thus, $KHCO_3$, for example, may be taken as a suitable compound to represent K in a carbonaceous matrix even though no H or C might occur in the matrix, but merely sufficient O or C to give the same or greater weight, in relation to the K present, as the weight of the HCO_3^- group in the compound $KHCO_3$.

Use of compounds rather than pure elements, apart from being necessary in certain cases, serves also to minimize second-order errors that might arise, for example, from differences in particle size, in bulk density, or in concentration range between the pure element and the sample as against the compound and the sample. With reference to the elements of interest above, the following compounds were chosen

Element	Representative compound
K	$KHCO_3$
Ca	CaO
Ti	TiO_2
Fe	Fe_2O_3

Correction for Matrix Background

The quantity R'' in the formulas above is obtained (as shown in the worksheets) from a quantity R by the formula

$$R'' = R - bR' \tag{4}$$

where R is the measurement for the given element in the sample and R' is the measurement of the sample for the radiation signal generated by the principal matrix material. The quantity b is the ratio of the values of R and R' obtained for a pure sample of the matrix material. Thus, the matrix material, in any quantity, gives a value of zero for R''. This condition may be represented by the equation

$$(FR) - \left(\frac{R}{R'}\right)(FR') = 0 \tag{5}$$

where F is a fraction related to the abundance of the matrix material.

In the present case, the matrix material was considered to have been made up of elements of atomic number 14 (Si) or less, e.g., Al, O, H. Since all such materials give a radiation signal of similar character, choice of any convenient compound that contained only elements of this type sufficed. The compound chosen for the above measurements was SiO_2.

Analysis of Calibration Sample

To calibrate the system for a given element, a sample of known concentration is required. This sample is used, along with a sample of the pure element or associated compound, to provide values for R'' and A to be substituted in Eq. 3, which is then solved for k.

The calibration sample may be analyzed by any one of various means, such as wet chemistry. However, the X-ray method itself may be used for such analysis by "spiking" a given sample with a known fraction of the element (or its representative compound) to be measured. The degree of spiking need not be restricted to small increments, and, in fact, it appears to be more accurate to use a large amount of spiking material and thereby to avoid the errors associated with the measurement of small differences.

The mathematical basis for the spiking procedure that was used is as follows. Assume that z grams of compound was added to g grams of unknown. Define the quantity q as

$$q \equiv \frac{z}{g+z} \tag{6}$$

The quantity q is thus a parameter that represents the increment in concentration of the compound in the unknown for the case of there having initially been zero quantity of the compound in the unknown. The actual concentration of the compound was initially the quantity p/C, and it may be shown that the actual increment in the concentration is the quantity $q[1-(p/C)]$ to give for the total concentration in the unknown the quantity $(p/C)+q[1-(p/C)]$. Solving Eq. 1 for k and substituting the latter quantity for p gives

$$k = \frac{(p/C) + q[1 - (p/C)]}{1 - \{(p/C) + q[1 - (p/C)]\}} \times \frac{A - R_d}{R_d} \quad (7)$$

where R_d, the output for the spiked sample, has been substituted for R'' (normally, R_d is made sufficiently large so that the matrix backgound is negligible, which is assumed to be the case here). Equation 3 can be restated as

$$p/C = \frac{1}{1 + \left(\frac{A - R''_u}{kR''_u}\right)} \quad (8)$$

where R''_u is the background-corrected output for the unspiked unknown. Equations 7 and 8 *could* be solved by substituting Eq. 7 into Eq. 8 and solving for p/C from the resulting quadratic relation in p/C. However, it is convenient to proceed as follows. Assume $p/C = 0$, solve Eq. 7 for k, and substitute in Eq. 8 to get p/C. Substitute this value of p/C in Eq. 7 to get a second value for k; substitute the second k value in Eq. 8, and test to determine whether the new value of p/C is substantially unchanged. If not, continue until this is so. This process converges very rapidly and is very easy to implement by use of a programmable desk calculator.

The method just described was used to determine Ti concentration in one of the unknown samples; TiO_2 was used as the representative compound.

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RADIOISOTOPES DATE THE "GENESIS ROCK"*

Supplementary Keywords: *geophysics; space study; activation analysis; activation, neutron; reactor; dating;* ^{39}Ar.

On the Apollo 15 mission, David Scott discovered a rock, identified as anorthosite in the Hadley Rille. The rock was dubbed the "genesis rock," because it is believed to be of the same material that formed the moon's original crust. The low abundance of radioisotopes in this moon rock—only $\frac{1}{20}$ that in most moon rocks—precluded the use of such means as the Rb/Sr and Pb/Pb methods to date it. Instead, Drs. L. Husain and J. F. Sutter, State University of New York at Stony Brook, used a different radioisotopic method. They irradiated 200 mg of the rock with fast neutrons in the high-flux reactor at the Brookhaven National Laboratory to convert ^{39}K to ^{39}Ar and then measured the $^{39}Ar/^{40}Ar$ ratio. They assigned to the rock an age of 4.15 ± 0.2 billion years, an age slightly less than the 4.5-billion-year estimate for the age of the solar system. Although the genesis rock is younger than some scientists had hoped, its age suggests that the lunar crust may have formed over hundreds of millions of years rather than as an almost instantaneous geological event. (HPR)

*From *Industrial Research*, p. 27, November 1971.

Cadmium Telluride Detectors

By F. Wald, R. O. Bell, and A. A. Menna*

(Summarized by Helen P. Raaen)

Supplementary Keywords: *chemistry, physical; source, alpha; source, gamma; apparatus; method; semiconductor; spectrometry; crystal; unusual condition; data handling;* ^{22}Na; ^{54}Mn; ^{60}Co; ^{137}Cs; ^{241}Am.

Abstract: *Cadmium telluride is especially suitable for general use as a gamma-ray radiation detector because its constituents have high atomic numbers. Tyco Laboratories has developed the traveling-heater method (THM) of growing single crystals of CdTe from a Te solvent; the crystals produced have low carrier concentrations and high mobility–trapping time products ($\mu\tau$). Tyco's continuing work has resulted in major progress: (1) reproducible synthesis and purification of CdTe by use of Te zones and r-f heating; (2) detailed analysis of the growth of CdTe by the THM, including the furnace construction, and of the results obtained; and (3) definitive characterization of CdTe detectors with respect to carrier concentration, gamma-ray detection, bulk inhomogeneity, and native-defect compensation. Fifty runs were made to synthesize or purify CdTe. The work is described in cogent detail—augmented by 36 figures, 8 tables, and the procedure for THM analysis—in USAEC Report NYO-4202-1, annual report for June 1, 1970–May 31, 1971; this article summarizes that report.*

In spite of their many advantages for use as detectors of gamma rays, semiconductors had a major disadvantage—restricted temperature range. The advent of high-resistivity CdTe removed this disadvantage, making possible their operation in the approximate range −150 to 150°C. The THM technique for the growth of single-crystal CdTe has resulted in detectors that have the necessary homogeneity, low trap density, and high resistivity. In this on-going improvement of the technique, crystal growth has been studied in depth to optimize CdTe preparation and to improve both crystal properties and growth parameters of the THM. Also, the CdTe products have been evaluated with respect to composition, performance, and modification.

CRYSTAL GROWTH

The principle of the THM crystal-growth procedure is shown schematically in Fig. 1. Compared with other techniques, it gives more nearly perfect crystals, controls composition more precisely, effectively purifies the source material, eliminates contamination from the crucible, and uses simple equipment that requires little attention. Experience with the technique has led to the design of the THM furnace shown schematically

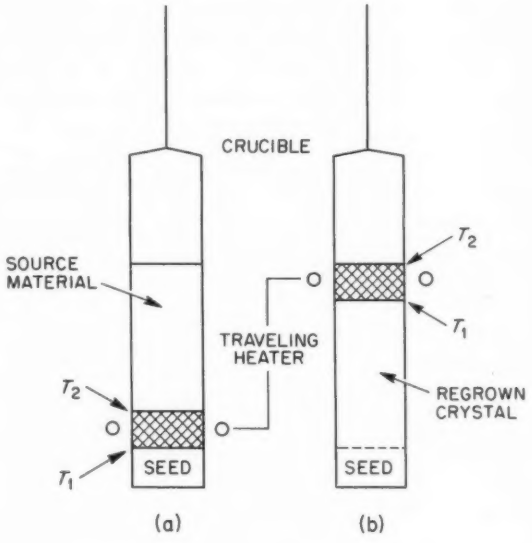

Fig. 1 Schematic of THM crystal-growth procedures (*a*) at beginning of growth and (*b*) at later stage.

in Fig. 2. These developments have resulted in prepurification of starting materials (obtained from the purportedly 99.999% pure semiconductors available commercially) to the very high purity of $\sim 10^{13}$ atoms/cm^3 carrier concentration. Repurification, by the THM, of the prepurified material gave standard starting materials of carrier concentration approaching that due to equilibrium amounts of native defects. By adjustment of the length of the solution zone, rate of ampul movement, solvent temperature, and temperature gradient at the interface, epitaxial crystal growth is controlled and optimized and occurs continuously.

The effects of several experimental variables on the CdTe product were evaluated in the 50 runs. Solvents

*Tyco Laboratories, Inc., Bear Hill, Waltham, Mass. 02154.

for the CdTe, including the most obvious one (Te), were studied—Bi, Cd, $CdCl_2$. The solvents Bi and Cd offered no advantages; the use of $CdCl_2$ will be explored further relative to Cl doping of CdTe. To

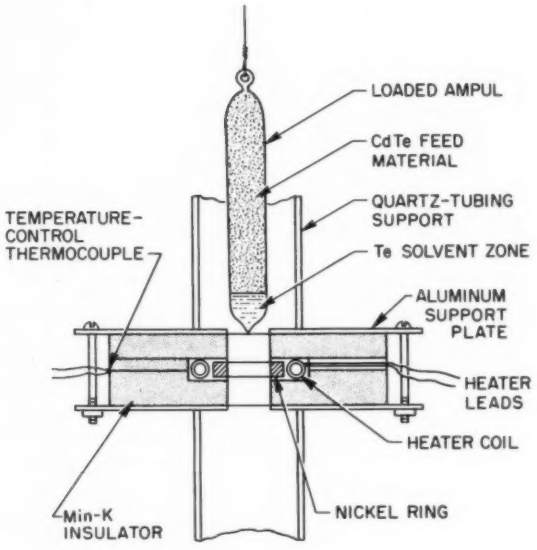

Fig. 2 Construction schematic of the THM-growth furnace.

prevent stress in the crystal, horizontal crystal regrowth (a zone-refining technique) was tried in addition to vertical regrowth but failed because of the high vapor pressure of Te. To obtain single-crystal growth for a sustained distance, modifications were made to the ampul, the temperature distribution in the solvent zone and interface was profiled, and seeding was attempted. A variation of the growth method—the traveling-solvent method (TSM)—was tried; it uses a thin zone and impresses a thermal gradient across the zone by heating the top slice and keeping the lower slice at a lower temperature. But growth over much distance was not sustained because of Te's high vapor pressure. The CdTe was intentionally doped with various elements, including Al, Cu, Dy, Fe, Ga, and In, by adding the impurity to the Te solvent zone. Except for Al and Dy, the dopants added could be backsegregated into the growing CdTe. Gallium and In acted as dopants to confer high resistivity on the CdTe. Copper was found to behave quite normally as described in the literature, and Fe produced essentially no change, probably because of lack of solubility in CdTe. Aluminum and Dy reacted with the quartz ampul and thus could not be properly studied, although the behavior of Al was qualitatively similar to that of Ga and In.

Temperature distribution during crystal growth by the THM is of interest relative to the speed and continuity of the single-crystal formation. Temperature distributions were measured experimentally (with difficulty and with dubious results!) and also were calculated with computer assistance. The computed distributions across (Fig. 3) and along (Fig. 4) the ingot and

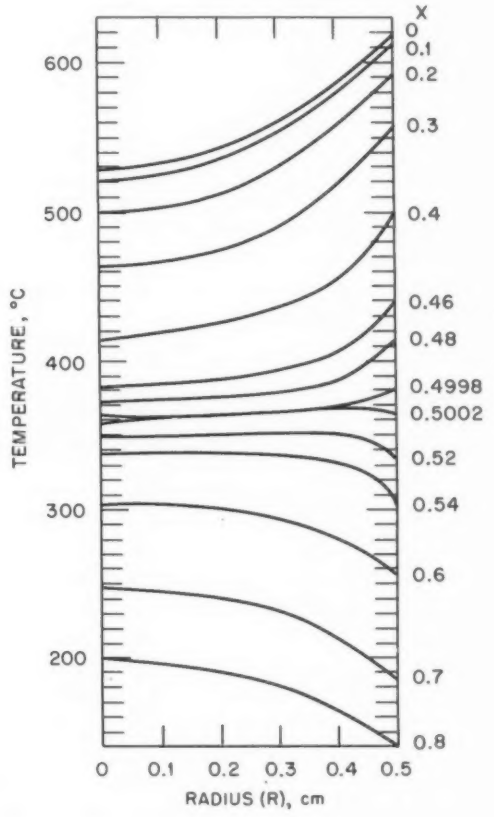

Fig. 3 Temperature as a function of the radius, R, for various values of the position along the ingot, X.

the calculated positions of the isothermal lines (Fig. 5) are especially interesting.

MATERIALS EVALUATION

At least three slices from each of the THM-grown CdTe ingots were used in evaluating the material.

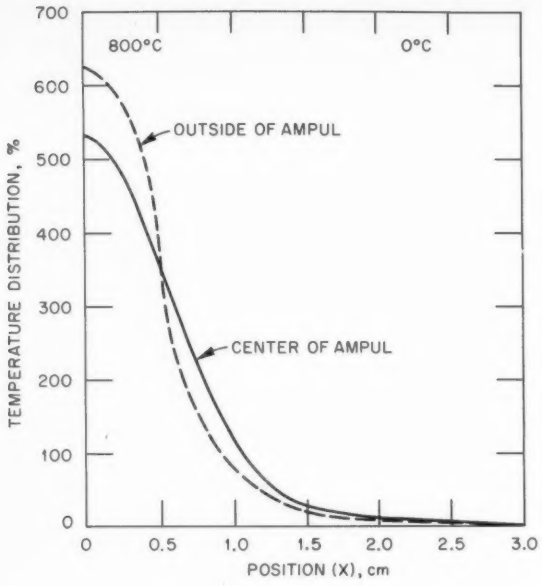

Fig. 4 Temperature distribution along the ingot at the center and on the outside of the ingot.

Carrier concentration was measured, optical transmission was observed, and a radiation detector was fabricated and used in radiation spectrometry.

The carrier concentrations were determined from measurements of the capacitance of a surface-barrier junction formed by pressure contacts with In, which is malleable, is an *n*-type dopant, and forms a rectifying barrier when it alloys with *p*-type CdTe. The carrier concentrations ranged from 10^9 to 2×10^{15} atoms/cm^3.

To detect bulk inhomogeneities (e.g., included Te) or voids, an infrared image converter was used. Figure 6 shows Te inclusions in a CdTe slice (an inclusion-free slice would be transparent). By transmission of far-infrared (2.5 to 40 μm) radiation, quality was also checked; for acceptable undoped material, no change in transmission occurs at wavelengths shorter than 25 μm. Some dopants produce a shift in the characteristic absorption.

The defect chemistry of the finished CdTe was also of interest. The effects of heat treatments and of compensation by doping on the deviations from stoichiometry in the CdTe products were explored experimentally. It was found possible to control carrier concentration by the very practical means of In doping at relatively high concentrations.

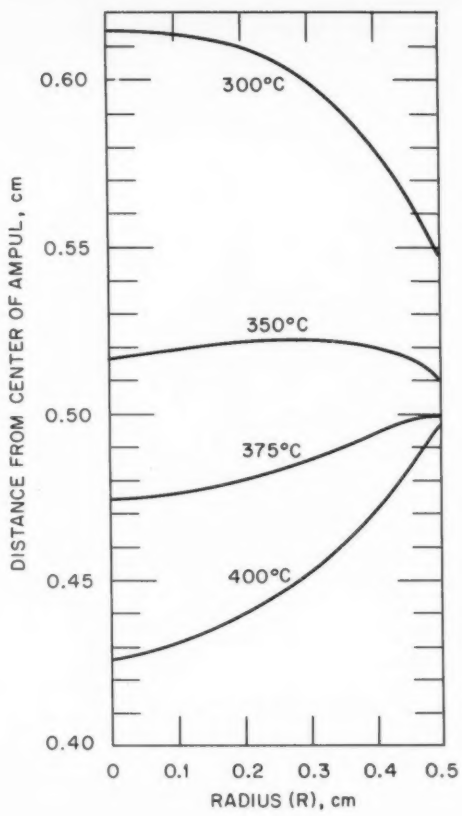

Fig. 5 Positions of selected isothermal lines as a function of radius and distance along the ampul.

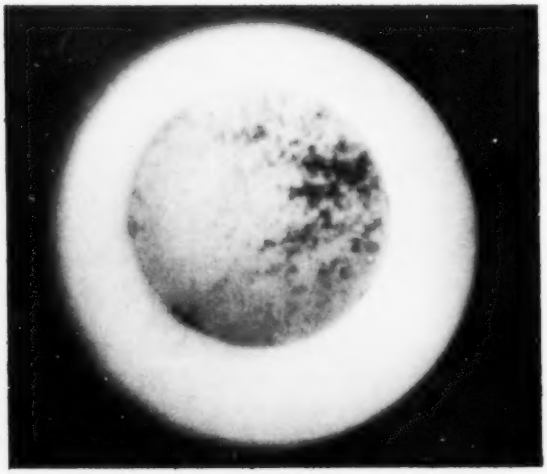

Fig. 6 Infrared optical transmission of a slice of THM-grown CdTe showing fine Te inclusions (dark areas).

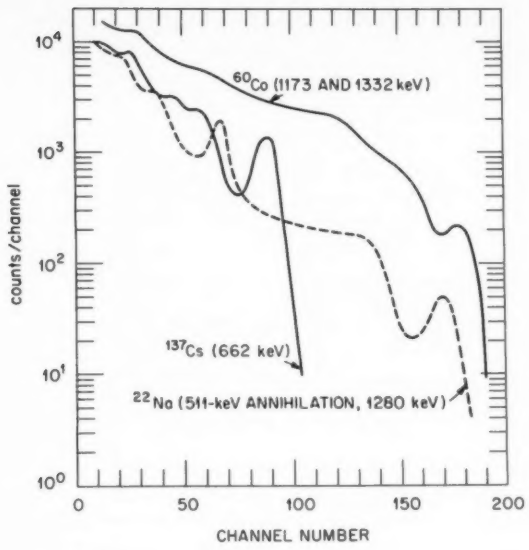

Fig. 7 Pulse-height spectrums of CdTe detector with ^{22}Na, ^{60}Co, and ^{137}Cs sources.

The surface barriers fabricated from the CdTe were evaluated as gamma-ray detectors with the gamma sources ^{22}Na, ^{60}Co, and ^{137}Cs; Fig. 7 shows the excellent resolution achieved. With the alpha-ray source ^{241}Am, the mobility–trapping time product ($\mu\tau$) was measured; this property was also excellent.

SUMMARY

The qualitative analysis of the THM has revealed a highly significant fact—the temperature peak in the middle of the solvent zone is the key feature in the THM process. It fulfills a very necessary function and must be preserved and maximized through proper design of the furnace. It allows flexible growth conditions by ensuring the proper balance between growth rate and temperature gradient, it avoids constitutional supercooling at the growth interface, and it aids diffusion through the solvent zone by providing convective currents that increase the effective material diffusion. The interface shape that seems most promising is a shape minimally convex toward the liquid.

^{2}H and ^{3}H in Studies of Heterogeneous Catalytic-Exchange Reactions

By Maria Frangopol*

Supplementary Keywords: *chemistry, organic; material tracing; reaction mechanism; isotope exchange; catalysis; review, technical; bibliography; enriched-stable-isotope use; ^{2}H.*

Abstract: *Uses of ^{2}H and ^{3}H as tracers in studies of heterogeneous catalytic-exchange reactions of hydrocarbons are reviewed. Reactions of alkanes and of cycloalkanes with ^{2}H, explained primarily by a dissociative mechanism, are given as examples. Studies on molecules with an asymmetric carbon atom, as well as on highly symmetrical molecules (methane, ethane, cyclopentane, -hexane, -heptane, -octane, and neopentane), are considered. Interaction of unsaturated hydrocarbons with the isotopes of hydrogen is a complex phenomenon that leads to hydrogenation, hydrogen exchange, double-bond migration, and* cis-trans *isomerization. Isotope exchange between aromatic hydrocarbons and ^{2}H on transition-metal catalysts is explained in terms of a π-complex mechanism. The associative and dissociative π-complex substitution mechanisms are compared with the corresponding classical mechanisms. Alkylbenzenes are discussed separately. Tritium-labeling methods based on isotope-exchange reactions are similar to those involving ^{2}H; several specific reactions are mentioned.*

Hydrogen isotopes are used in two ways to study the mechanism of catalytic-exchange reactions. One consists in following experimentally the distribution of the tracer isotope among various positions in the molecule of a particular starting material, reaction product, or both. Thus information is provided on the relative reactivities of the atoms in the different selected positions. The second is more complex and requires the determination of the distribution of tracer isotope among the molecules of reactants and products. This method gives data on the role of the labeled substance

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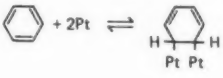
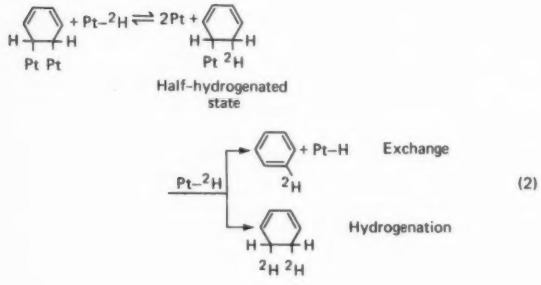
in a multistage catalytic process, the properties of the catalyst, and the nature of the products. The combination of isotope-tracer studies, information on structural variation, and stereochemistry has been particularly useful in testing the chemical mechanisms of heterogeneous catalytic reactions of hydrocarbons.

ISOTOPE EXCHANGE BETWEEN AROMATIC HYDROCARBONS AND ^2H

The classical mechanisms proposed for catalytic-exchange reactions of aromatic hydrocarbons are associative and dissociative exchange mechanisms.[1] The main feature of the associative theory advanced by Horiuti and Polanyi[2] is that the unsaturated hydrocarbons chemisorb by the opening of a double bond

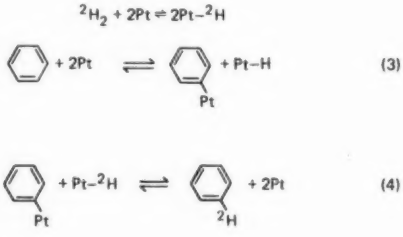


(1)



(2)

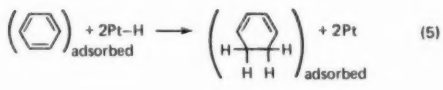
The dissociative mechanisms proposed by Farkas and Farkas[3-5] explain that hydrocarbon molecules chemisorb through C–H bond rupture



(3)

(4)

Contrary to the associative mechanism, which explains that both exchange and hydrogenation occur on the same path (Eq. 2), the classical dissociative theory proposes that they occur by two unrelated mechanisms. Exchange takes place by the mechanism given in Equations 3 to 5, and hydrogenation involves the simultaneous addition of two hydrogen atoms, chemisorbed on the catalyst, to the hydrocarbon physically adsorbed in the Van der Waals layer of the catalyst



(5)

Arguments exist both for and against the dissociative mechanism.[1] The strongest counterevidence is that exchange reactions of unsaturated hydrocarbons are very much faster than those of saturated hydrocarbons.[6,7] This observation is difficult to explain by the dissociative mechanism, which—because electrons are not involved in the dissociative process—predicts approximately similar reactivities for saturated and unsaturated hydrocarbons under conditions where reagent displacement and poisoning side reactions are negligible. On the other hand, the fact that exchange reactions occur between normal and deuterated hydrocarbons favors the dissociative mechanism.[1]

But along with these evidences in favor of one or the other mechanism are some limitations and difficulties associated with classical theories. The chief ones are (1) the interpretation of the differences in adsorption strengths of chemically similar aromatic hydrocarbon molecules (benzene, diphenyl, naphthalene, anthracene, etc.) observed during exchange[6,8,9] and hydrogenation reactions;[10] (2) the occurrence of randomization reactions; and (3) the rapid exchange rate of unsaturated hydrocarbons.

A new approach was thus necessary to explain the difference in the adsorption strengths of aromatic hydrocarbons in such a manner as to involve the different electron properties of the various molecules.[11] This approach has been interpreted in terms of a third mode of adsorption, whereby unsaturated hydrocarbons are adsorbed on Group VIII transition metals by π-complex formation.[8,9] As a result, two new exchange mechanisms were formulated on the basis of π-complex adsorption—the associative and dissociative π-complex substitution mechanisms. Related to these approaches, new mechanisms can also be advanced for hydrogenation and isomerization reactions.

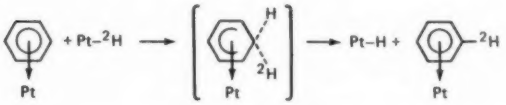
The π complexes are described in terms of Mulliken's charge-transfer theory,[12,13] which attributes the complex-forming reactions of stable aromatic molecules to an electron donor–electron acceptor interaction. The stability of a π complex involves not only the

π-electron donation into empty *d* orbitals of the metal but also the significant back-donation of metal *d* electrons into vacant antibonding orbitals of the olefin.[14] The direction of net charge transfer is explained quantum-mechanically by symmetry factors associated with the bonding and antibonding orbitals of the organic molecule. Since antibonding orbitals are more complex and contain more nodal planes than do bonding orbitals, the bonding-orbital interaction should predominate so that net charge transfer occurs toward the metal, as in the representation

I

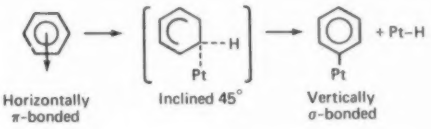
The new mechanisms postulated on the basis of π-complex adsorption for transition-metal-catalyzed exchange reactions between aromatic molecules and water or hydrogen gas[8,9] differ from their classical counterparts in that the dissociative mechanism involves electrons in the dissociation process, whereas the opening of a double bond is not involved in the new associative mechanism.[11]

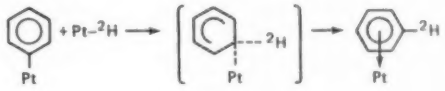
The associative mechanism is comparable to a conventional radical (hydrogen atom) substitution reaction wherein the π-bonded benzene molecule is attacked by a hydrogen atom formed by the dissociative adsorption of water or hydrogen gas. The activation energy in this process results from the partial localization of one π electron in the transition complex,[15,16] and the transition state is π-bonded to the catalyst surface



A characteristic of the transition state is its formal resemblance to the "half-hydrogenated" chemisorbed intermediate of the classical associative mechanism previously discussed. Because of the short lifetime of the transition state, the smaller π-electron localization, and the lack of dependence on lattice spacings, this transition state is not affected by the same energy restriction as the half-hydrogenated intermediate.

Dissociative π-complex substitution mechanism implies that the π-complex–adsorbed aromatic compound reacts with a metal radical (active site) by a substitution process. The molecule rotates through 90° and changes from its horizontal π-complex–adsorbed position to a vertical σ-bonded chemisorbed state





Concerning the second reagent in the exchange reaction, namely, ^2H_2O or $^2H_2$ ($^3H_2$) gas, the rapid exchange on transition-metal catalyst can be explained by dissociative chemisorption

$$^2H_2 + 2Pt \rightleftharpoons Pt\text{–}^2H + Pt\text{–}^2H$$
$$H_2O + 2Pt \rightleftharpoons Pt\text{–}H + Pt\text{–}O\text{–}H$$
$$Pt\text{–}O\text{–}H + Pt \rightleftharpoons Pt{=}O + Pt\text{–}H$$

Various catalytic-exchange reactions with benzene[17-19] and with other aromatic hydrocarbons[20] have been reported, but the most useful general one-step method for labeling aromatic compounds with ^2H, ^3H, or both is the heterogeneous platinum-catalyzed exchange with isotopically labeled water.[21]

Gamma rays from ^{60}Co and ultraviolet light from a low-pressure source were used to study the effect of preirradiation in the exchange between benzene and $^2H_2$ in the presence of self-activated platinum oxide.[22] Under these conditions, the rates of both multiple and stepwise exchange processes were accelerated. Of 14 oxides examined, including those of the Group VIII transition metals, platinum oxide was the only one to catalyze exchange at temperatures up to 130°C. Preliminary experiments indicated that radiation-induced desorption of oxygen from the surface of the oxide was responsible for the accelerative effect. Internal radiation sources, such as ^3H_2O, also increased initial exchange rates. Naphthalene, diphenyl, benzoic acid, cholesterol, and diphenylether were studied with ^3H_2O, and the results were rationalized in terms of π-complex mechanisms for exchange.

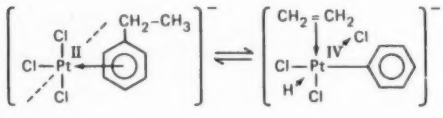
Some controversy now exists as to whether heterogeneous exchange of aromatic hydrocarbons occurs through associatively or dissociatively adsorbed intermediates.[11,23,24] Since the associative and dissocia-

tive mechanisms of π-complex substitution are not mutually exclusive, both may participate simultaneously in exchange reactions where ^2H_2O is the second partner. However, it is well established that the exchange reaction between a normal and a deuterated aromatic hydrocarbon occurs through a dissociative mechanism.[25]

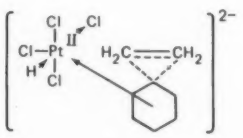
ISOTOPE EXCHANGE BETWEEN ALKYLBENZENES AND ^2H

The catalytic-exchange reaction of hydrogen atoms in alkylbenzenes in the presence of different metals has been studied.[26] Initially, the exchange of the ring hydrogens was explained on the basis of an adsorbed state,[27] account being taken of the geometric factors proposed by Twigg and Rideal,[28] and later, by means of π complexation. Detailed studies on the exchange reaction between ^2H and toluene,[29] and between ^2H and *p*-xylene, ethylbenzene, *tert*-butylbenzene, and *p-tert*-butyltoluene[23] have been reported. The isotopic "isomerization," or self-exchange, reaction of ^2H in monodeuterotoluene has been investigated with its three species—that is, deuterated at the *ortho*, the *meta*, and the *para* positions;[30] mechanisms have been proposed for both exchange[9,11,26,31] and hydrogenation[32,33] reactions.

Homogeneous and heterogeneous metal-catalyzed isotopic hydrogen exchanges in alkylbenzenes have been compared.[34] In homogeneous platinum-catalyzed exchange, the isotope incorporation in alkylbenzenes occurs in both ring and side-chain positions.[35,36] In ethylbenzene, the α and β positions exchange at almost equal rates. Mechanisms considered[36] to account for this orientation include symmetric intermediates involving reversible dealkylation



and nonclassical carbonium ions, such as the π-bonded ethylene phenonium ion (II).



II

The validity of these mechanisms was an important question not only for fundamental catalytic theory but also for its practical implication in ^2H and ^3H labeling of the alkylbenzenes. The occurrence of reversible dealkylation would severely limit the use of the technique of homogeneous exchange, particularly for tritiation,[34,37] since chemical decomposition as a consequence of dealkylation could yield significant quantities of impurities, which would prejudice ultimate radiochemical purification of the compound. Experimental results show that the above mechanisms do not apply; they favor an alternative mechanism that involves π-allylic species.[36]

In the heterogeneous platinum- and nickel-catalyzed exchange of β-^{14}C-ethylbenzene, radioactivity measurements showed that dealkylation does not occur. A process involving adsorbed π-allylic intermediates was considered as a possible mechanism for isotope incorporation.[38,39] Ethylbenzene becomes deuterated more extensively in the α position of the side chain than in the corresponding β position, especially when compared with the homogeneous analog. This finding suggested that an additional surface mechanism is operating only in the heterogeneous system; it was thought to be the abstraction process proposed by Garnett and Sollich-Baumgartner.[40]

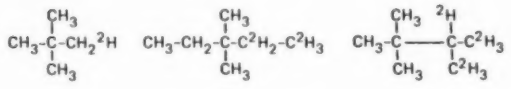
ISOTOPE EXCHANGE BETWEEN ALKANES AND ^2H

Some arguments of Farkas and Farkas for the dissociative mechanism of exchange result from experiments on the exchange of ^2H with saturated hydrocarbons.[4] Saturated hydrocarbons do exchange with ^2H and also exchange ^1H atoms among themselves.[41] This statement was first made by Morikawa, Benedict, and Taylor[42] and by Morikawa, Trenner, and Taylor.[43] Later, Farkas and Farkas[4,44] studied the exchange of ethane, propane, butane, *n*-hexane, and cyclohexane on active-platinum catalyst.

Isotope-exchange reactions between alkanes and ^2H are reviewed by Taylor,[1] by Anderson,[45] and by Gelbin and Wilde;[46] the associative and dissociative mechanisms are discussed primarily. However, there now seems to be little disagreement with the consensus that a dissociative mechanism explains the exchange reactions of saturated hydrocarbons.

The initial products of the exchange of normal alkanes and methylalkanes at 0 to 200°C contain all exchanged species between alkane-d_1 and $C_n{}^2H_{2n+2}$. Those of certain other hydrocarbons contain no species

more exchanged than compounds III and IV (Ref. 47). Interesting results concerning the barrierlike behavior



III
Neopentane[48,49]

IV
3,3-Dimethylpentane[50,51]

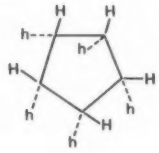
V
2,2,3-Trimethylpentane[52]

of a quaternary carbon in the molecule have been obtained on 3,3-dimethylhexane and on 2,2,3-trimethylbutane.[53]

A major problem of continuing interest in heterogeneous catalysis—the influence of surface geometry on the mechanism of the reaction—was investigated by means of the saturated hydrocarbons. To assess the influence of surface structure on the mode of adsorption of alkanes, ^{2}H-exchange reactions with propane, ethane, and methane were studied over various ultra-high-vacuum-deposited nickel films.[54]

ISOTOPE EXCHANGE BETWEEN CYCLOALKANES AND ^{2}H

The hydrogen atoms of cyclopentane have been divided arbitrarily into two sets, H and h, to explain the mechanism of the exchange reaction; each set consists of all hydrogen atoms on one side of the ring



VI
Cyclopentane

Burwell and Tuxworth[55] used the term "interchange reaction" to define the step that permits the propagation of the exchange from the first set of hydrogen atoms to the second. The mechanisms and different formulas for the intermediates have been proposed.[47,51,56]

Isotope exchange of hydrogen in cyclohexane in the presence of various catalysts, e.g., nickel,[57] films of partially poisoned evaporated platinum,[58] and zeolites,[59,60] has been investigated. Also, studies have been made of exchange in cyclobutane;[61] in cycloheptane and cyclooctane;[62] in cyclopentane, bicyclo[3.3.0]octane, bicyclo[3.2.1]octane, bicyclo[3.3.1]nonane, and cyclopentylcyclopentane;[50] in 1,2-dimethylcyclohexane and decalin;[63] and in 1,2-dimethylcyclohexane.[64,65] Since stereochemical resemblances between racemization and two-set exchange in cyclopentane have been noticed, a single mechanism that could account for both processes has been sought.[51,55]

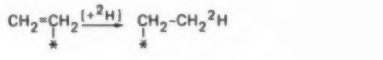
REACTION OF OLEFINS WITH ^{2}H

In one of the earliest works on the reaction of ^{2}H with olefins on nickel catalysts, Farkas, Farkas, and Rideal[66] found that ^{2}H not only adds to the double bond but also exchanges with the ^{1}H of ethylene. It is now known that ^{2}H–olefin reactions which occur on the catalyst surface under hydrogenating conditions usually involve chemisorption on the catalyst surface, addition to the double bond, redistribution of ^{1}H and ^{2}H in the molecules, exchange between olefin and ^{2}H, exchange between olefin molecules, double-bond migration, *cis–trans* isomerization, self-hydrogenation and decomposition, dehydrogenation, and polymerization.[1]

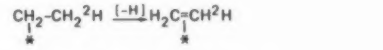
The adsorbed state of olefins and the interaction between olefins and catalyst have been much discussed.[67-70] The formation of an adsorption complex on the surfaces of transition metals has been proved by the increase of the surface potential of the metals[71] and seems to explain the mechanism of the exchange between ^{2}H and olefins.

Sigma-adsorbed olefins[72] along with π-adsorbed olefins have been considered in attempts to establish various mechanisms. Particularly for α-methyl olefins, π-allyl complexation in *syn* and *anti* configuration has been proposed.

The mechanism of the olefinic exchange is based on the addition of a ^{2}H atom to the π-adsorbed olefin; the addition leads to a monodeuteroethyl radical



where * is the catalyst. The process of the olefinic exchange is then realized by elimination of a ^{1}H atom followed by the desorption of the olefin



Much experimental information has been obtained by many authors on the metal-catalyzed reaction between olefins and ^{2}H (Refs. 1, 33, and 73).

Ethylene has been hydrogenated with ^2H on the weaker and stronger active sites of alumina, which had been found previously with hydrogen to give different activities and kinetics for the reaction. It has been reported[74] that on the weaker sites at low temperatures, the main product is 1,2-ethane-$^2H_2$ with a small amount of ethane-$^2H_3$, whereas on the stronger sites at higher temperature, H–^2H exchange of ethylene occurs much more quickly than hydrogenation, and all deuteroethanes are produced. The results for the weaker sites have been explained according to a Twigg–Rideal mechanism. For the reaction on the stronger sites, a different mechanism was proposed that involves reaction of ethyl carbonium ions with hydrogen atoms formed by the dissociative adsorption of hydrogen on the weaker sites.

The reactions of ethylene and of other olefins with ^2H have also been studied on chromic and zinc oxides[75] and on titanium dioxide.[76]

Propylene and ^2H_2O rapidly exchange ^1H against ^2H at 360°C in the presence of SnO_2 covered with MoO_3 (Ref. 77). The distribution of the ^2H over the propylene molecules proved to be approximately Gaussian. One of the ^1H atoms in the propylene molecule—supposedly one of those bonded to the central atom—participated only slightly in the exchange. Infrared absorption measurements, confirmed by proton magnetic resonance, revealed that ^2H appears preferably in the methylene group and, for the greater part, in the *cis* position with respect to the methyl group.

The ^2H-exchanged position of the monodeuteropropylene produced in the exchange reactions of propylene on Cu, Rh, Ni, Pd, and Pt was investigated by microwave spectroscopy.[78,79] Results showed that the adsorbed propylene species differs drastically according to metal and that double-bond migration occurs during the adsorption and desorption.

Perdeuteroolefins from C_2 to C_5 having isotopic purities >98% were prepared by a ^2H-exchange reaction over a pure alumina catalyst at room temperature.[80] The mechanism of the reactions of 1-butene, *cis*-2-butene, isobutene, and *tert*-butylethylene with ^2H has also been investigated; only vinyl C–H bonds undergo exchange below 100°C (Ref. 80). The exchange occurs most readily with terminal vinyl hydrogen atoms of α olefins. The exchange is postulated to occur by a dissociative mechanism at acid–base or ion-pair sites on the dehydroxylated surface.

The relative rates of exchange, addition, and double-bond migration of 1-butene on a nickel-wire catalyst were studied.[81] The double-bond migration has been explained in terms of an alkyl- or allyl-radical intermediate.[72]

Studies on higher olefins, e.g., 1-hexene, have been reported,[82] and papers exist[83,84] that are devoted to the cycloolefins.

ISOTOPE-EXCHANGE REACTIONS WITH ^3H

The isotopes of hydrogen have been used extensively in attempts to elucidate the mechanism of exchange reactions.[85,86] Along with ^2H, ^3H (Ref. 37) has contributed significantly in this field. General methods for labeling organic compounds with ^3H have been reviewed,[87] and mechanisms and intrinsic limitations of direct processes have been discussed.[88] Garnett pointed out the potential of heterogeneous catalytic exchange as a ^2H- and ^3H-labeling method[21] for use in organic syntheses; he applied π-adsorption complex theory to predict total isotope uptake and isotope orientation within a given molecule.

Many resemblances exist between the exchange reactions of hydrocarbons with ^2H and with ^3H, but the β-emitting characteristics of ^3H (Ref. 89) and the isotope effects[1] must be considered.

Specifically, ^3H was used in exchange reactions with alkanes,[90-93] cycloalkanes,[94,95] olefins,[93,96,97] and aromatic hydrocarbons.[98-100] (HPR)

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Single-Crystal High-Resistivity Cadmium Telluride

By K. R. Zanio*

(Summarized by Helen P. Raaen)

Supplementary Keywords: *source, alpha; source, gamma; industry, electronics; physics; dosimetry; method; secondary radiation effect; backscatter; semiconductor;* ^{57}Co; ^{60}Co; ^{85}Sr; ^{113}Sn; ^{137}Cs; ^{212}Bi; ^{212}Po; ^{241}Am.

[Editor's Note: At Hughes Research Laboratories, research sponsored by the Division of Isotopes Development (DID) is progressing steadily to develop cadmium telluride (CdTe) semiconductor detectors for use in room-temperature gamma-ray spectrometry. The status of this work is described in the annual report summarized here and in several other publications.[1-10] When these devices become available commercially, they will be the means to several new applications of radioisotopes, especially in medical diagnostics and in mineralogy.]

Abstract: *Single crystals of semi-insulating CdTe were grown from the vapor and by a modified Bridgman technique. Surface-barrier detectors made from them resolved photopeaks from monochromatic X- and gamma-ray emitters in the energy range 59.6 keV (^{241}Am) to 1.33 MeV (^{60}Co). The detectors demonstrated an increase in charge-collection efficiency from ~50% (Bridgman) and ~80% (vapor) to ~95%. With an ~15-mm³-active-volume detector, a 9:1 peak-to-volume ratio was attained for ^{57}Co (122 keV). Another detector of similar size gave 9-keV resolution with 59.6-keV X rays. The achievement of an increase in the homogeneity of the crystals accounts for the improvement in the response of the detectors made from them. This article is a summary of USAEC Report SAN-549-7, Annual Report for Jan. 1–Dec. 31, 1970.*

For room-temperature gamma-ray spectrometry, germanium and silicon (common semiconductor materials) are unsuitable. Germanium must be cooled during use because its narrow bandgap (0.68 eV) causes excessive thermally generated currents. Silicon's low atomic number (14) results in a predominance of Compton processes in the gamma-ray interactions. Cadmium telluride was believed to be the best candidate because it has a high atomic number (~50), wide bandgap (1.5 eV), and promising transport properties.

Earlier work at Hughes Research Laboratories showed that CdTe can resolve photopeaks from monoenergetic gamma emitters. With Bridgman-grown crystals, the results could be obtained only at a reduced charge-collection efficiency (~50%) because of the poor transport properties of holes. With vapor-grown material, photopeaks were obtained at an improved charge-collection efficiency (~80%) due to improved material homogeneity. In both cases, however, the peak-to-valley ratio was <2:1.

The techniques for growing single CdTe crystals were improved. As a result, crystals having greater homogeneity were obtained. Spectrometers fabricated from them perform better and now have possible practical application.

CdTe IN ROOM-TEMPERATURE GAMMA-RAY SPECTROMETRY

For room-temperature spectrometry of low-energy gamma rays, the resolving power of devices made from semi-insulating CdTe has become comparable with that of NaI scintillation. Figure 1 shows the ^{57}Co spectrums for NaI and a CdTe device fabricated from vapor-grown material. For the CdTe and NaI detectors, respectively, the resolutions [full-width half-maximum (FWHM)] are 16 and 14%, and the peak-to-valley ratios are 9:1 and 25:1. For the CdTe device the total charge-collection efficiency is 95%. The gamma-ray spectrums of ^{60}Co (1.17 and 1.33 MeV), ^{113}Sn (0.393 MeV), ^{85}Sr (0.513 MeV), and ^{137}Cs (0.662 MeV) and the X-ray spectrum of ^{241}Am were also used to evaluate the CdTe devices.

PRECIPITATES AND TRAPS— DEGRADERS OF RESOLUTION

Incomplete charge collection degrades the gamma response in CdTe. When gross imperfections and traps intercept the drifting electrons and holes that have been created by ionizing radiation, total charge collection cannot be attained. Gamma spectrums reflect this charge-transport process. The ideal detector can reproduce, through appropriate electronics, the energy spectrum deposited in it. Since all charge will be collected at the electrodes in the ideal detector, the

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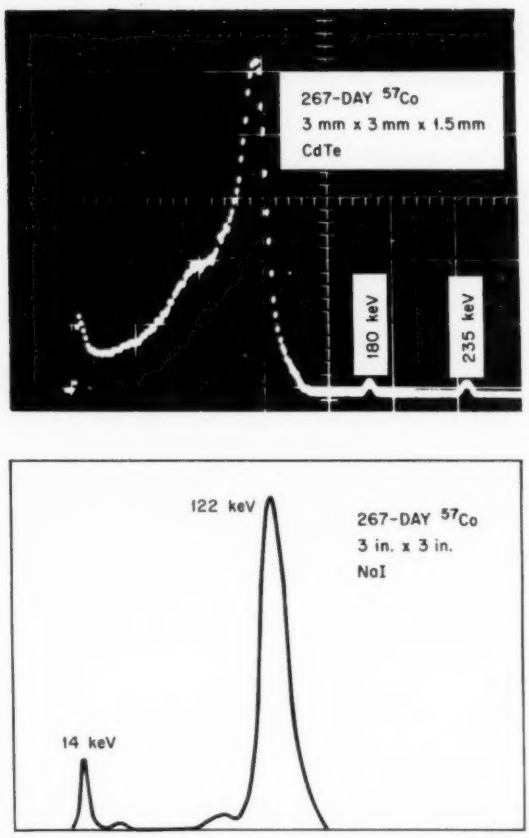

Fig. 1 Room-temperature gamma spectrums taken with vapor-grown semi-insulating CdTe (top) and NaI (bottom) using ^{57}Co (122 keV).

magnitude of the pulse will be the same regardless of where the electron–hole pairs are created. The pulse-height distribution is a spike and corresponds to 100% charge-collection efficiency for both electrons and holes. Figure 2 presents diagrammatically the ideal and degraded spectrums for photoevents and the corresponding states of the detector with respect to traps and gross imperfections.

Precipitates

In CdTe crystals, the gross imperfections are most likely due to precipitates (Fig. 3) that result from the nature of the Cd–Te phase diagram. In both the Bridgman- and vapor-growth techniques, crystals were grown in the region of the phase field where precipitation is minimal. The larger precipitates ($\sim 5\,\mu$) have been removed or reduced in size by thermal annealing;

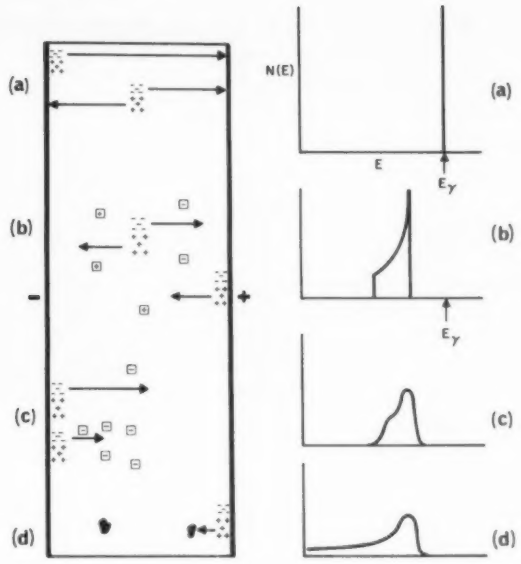

Fig. 2 Gamma spectrums associated with a detector containing traps and gross imperfections (*a*, ideal; *b*, traps; *c*, fluctuation in density of traps; *d*, gross imperfections).

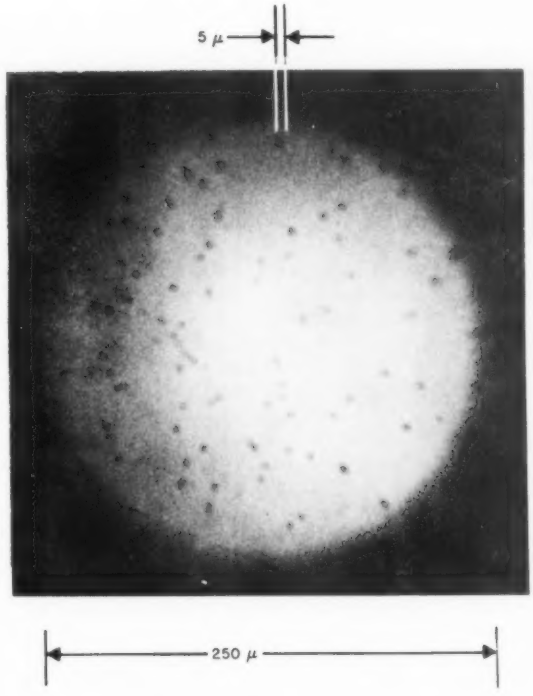

Fig. 3 Infrared photograph of 5-μ imperfections that degrade resolution in semi-insulating CdTe.

infrared microscopy was used to detect them. Submicron precipitates and their effect on spectral resolution were studied by means of the spectrums of alpha particles from ^{241}Am. An analysis of the spectral response of a surface-barrier detector to alpha particles provides information regarding the submicron precipitates, since they also intercept the charge cloud of electrons or holes as they traverse the crystal.

Traps

Trapping is caused by native defects, foreign atoms, or the association thereof in the crystal; Fig. 4 is a diagram of the process. In the case of deep trapping,

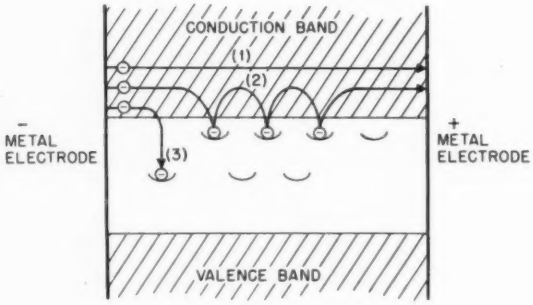

Fig. 4 Transport of electrons within a surface-barrier device for (1) no trapping, (2) shallow trapping, and (3) deep trapping.

carriers are trapped but not detrapped. As a result, total charge collection cannot occur. These are the trapping centers that degrade the performance of CdTe as a room-temperature spectrometer. An analogous transport process pertains to holes injected at the positive electrode. Because of the interplay between foreign atoms and native defects, a change of resistivity results in a change in the position and density of trapping sites. The transient-charge technique is the most useful tool to analyze the trapping phenomenon when the material is in its semi-insulating state as a gamma detector. An analysis of the transient response by use of a pulsed electron beam is similar to that in which alpha particles are used, except that more precise data are obtained because signal-to-noise ratios are better. Analysis of CdTe with this technique has provided:

1. Comprehensive measurement of the electron drift velocity.

2. Measurement of trapping time for electrons as a function of electric field.

3. Correlation of drift velocity with trapping time.

Figure 5 shows the drift-velocity data.

CRYSTAL GROWTH

Single crystals have been grown at Hughes by both vapor and Bridgman techniques. A boule of CdTe

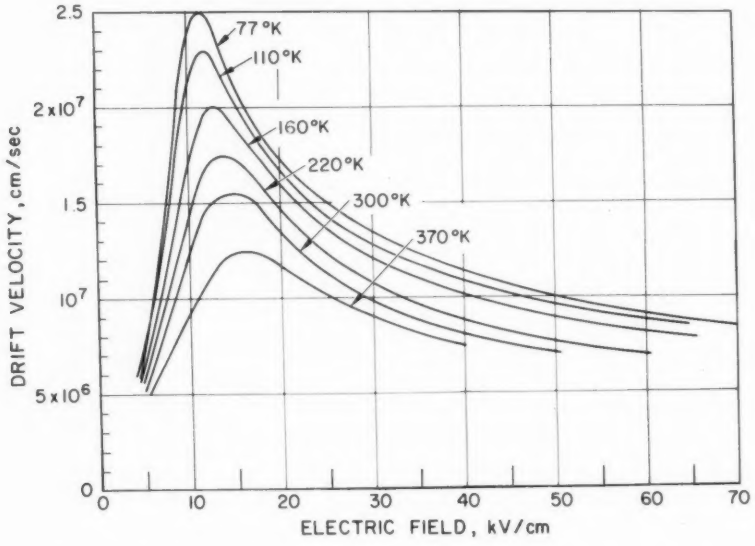

Fig. 5 Drift-velocity characteristic for electrons in semi-insulating CdTe.

(vapor-grown, 90% crystal) is shown in Fig. 6. Single crystals up to 3 cm^3 in volume were achieved. This upper value is over an order of magnitude larger than previously reported. An increase in the crystal size is due to control of the direction of crystal growth with thermal gradients. At present, the typical size of vapor-grown crystals is ~1 cm^3, which is adequate for spectrometry. The improved spectral resolution attained with the devices made from this more recent material results from an improvement in the homogeneity of the single crystal. However, no increase in the mobility–trapping-time product ($\mu\tau^+$) was achieved, and the products were not significantly different for the vapor- and Bridgman-growth techniques. The best values in the same single crystal are 2×10^{-4} cm^2/V for electrons and 4×10^{-5} cm^2/V for holes.

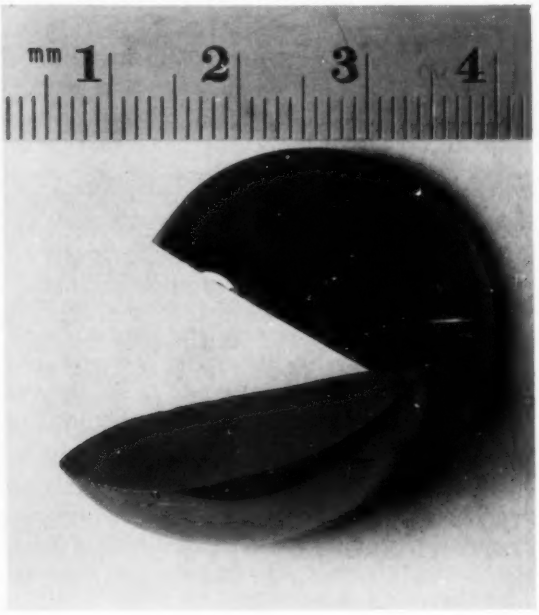

Fig. 6 Boule of vapor-grown semi-insulating CdTe, of which 90% is single crystal.

Large (20 cm^3) single crystals of semi-insulating CdTe had been obtained at an early date by the Bridgman technique. Recently, improved homogeneity has been obtained with this method by extracting heat axially during growth. To help transfer heat along the crystal and to establish a convex (pointing in the direction of growth) or at least a planar crystal-melt interface, a lowering mechanism was used that supports the bottom of the crucible and consists of a metal rod driven by a lead screw mechanism. This type of support provides a heat sink on the end of the crystal to direct the heat axially down through the crystal.

CONCLUSIONS

The room-temperature performance of gamma-ray spectrometers made from semi-insulating CdTe has been improved significantly. Improved photopeak-to-valley ratios permit superior discrimination from Compton events and from photons emitted from other isotopes. These factors are especially relative to nucleonic gaging where ~500-keV-gamma emitters are considered. Because of the feasibility of room-temperature operation and the high photoelectric absorption coefficient at 100 keV, CdTe devices are promising in arrays that might be used in a gamma camera for medical diagnostics. The best peak-to-valley ratios attained thus far [9:1 with ^{57}Co (122 keV)] are more than adequate for this application. At lower energies, this material has promise as an X-ray detector [9 keV (FWHM) at 59.6 keV].

At present, the active volumes of the devices are restricted (10 to 15 mm^3) because charge collection is restricted by trapping and precipitates. Transient-charge techniques, alpha-ray spectrometry, and infrared microscopy are being used as tools to characterize these imperfections. Further refinement in the crystal-growth process is necessary to decrease the precipitates and to minimize the trapping.

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Principles and Techniques of Alpha-Particle Gaging

By E. J. Driscoll, T. E. Crume, K. Verghese, and R. P. Gardner*

The fact that alpha particles have not been used as much as gamma radiation and beta particles for gaging purposes can probably be attributed, at least in part, to two major technological problems that have now been essentially solved: (1) obtaining a suitable detector for alpha particles and (2) obtaining suitable matrices for producing stable and rugged source configurations for alpha-particle-emitting radioisotopes of practical half-lives.

The first of these problems has been solved by the advent of thin surface-barrier-type silicon detectors that are capable of detecting alpha particles with nearly 100% efficiency while exhibiting almost 0% efficiency for gamma radiation and beta particles. In addition, they are capable of spectrometry, have negligible window thicknesses, and can be operated at normal temperatures and pressures.

The second problem has been alleviated by advances in alpha-particle source technology over the past decade to the point that matrices are now available which are resistant to abrasion, temperature changes, and exposure to water. In particular, ^{241}Am can now be obtained as an essentially windowless sealed source in quantities exceeding 1 mCi. This radioisotope has a half-life of about 460 years and a primary alpha-particle energy of 5.48 MeV.

These advances in alpha-particle detection and source technology have already contributed to an increase in interest in the use of alpha particles for gaging. It is likely that this interest will increase even further in the future as alpha-particle radiation offers the possibility of several unique measurement principles and ranges of measurement.

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The present article reviews the state of the art of alpha-particle gaging. The review is organized on the basis of principles of measurement, and the various applications of these principles are discussed in each category. We have not attempted to cite all references on this topic; rather we have attempted to select references that cover specific points of interest. We would like to acknowledge the help of the Isotopes Information Center at Oak Ridge National Laboratory in providing many of these references.

PRINCIPLES OF ALPHA-PARTICLE GAGING

Alpha-particle gaging can be conveniently categorized on the basis of (1) transmission, (2) elastic scattering, and (3) interaction effects. There are also a number of subcategories that can be identified for each. These will be discussed, and relevant research and development work will be described.

Transmission

Transmission principles essentially involve an alpha-particle source separated from a suitable detector, with the sample under investigation interposed between them. The sample may be a thin film or a gas. The detector response is then obtained as a function of a sample property—e.g., film thickness, gas density, gas temperature, gas pressure, or gas composition. Subcategories, based on the type of detector used, include range, residual energy, and energy shift.

Range. The range transmission principle is based on detector systems that simply count each alpha particle which reaches them and gives equal weight to each, regardless of energy—e.g., G–M counters or

pulse-type ion chambers, proportional counters, or solid-state detectors operated simply as counters. Since thin and thick alpha-particle sources give rise to marked differences in response, they are discussed separately.

Thin Sources. Thin (monoenergetic) sources can be used to give high sensitivity over a very narrow range of sample properties by taking advantage of the alpha-straggling phenomenon which, for point sources and detectors, allows very accurate measurement of sample properties over a range equivalent to about 1% of the total range of the alpha particles. The primary problem associated with this principle involves adjusting the range of measurement. For film thickness, this has been done by adjusting either the source–detector spacing or the ambient gas pressure. The former method has the disadvantage that the response intensity varies with the change in geometry as well as with the film thickness so that a calibration must be made for each source–detector spacing. This technique has also been used for adjusting the range of measurement when the properties of gaseous samples are being gaged, but again suffers from the same disadvantage.

Barber,[1] Enge and Wahlig,[2] and Ramavataram and Porat[3] developed alpha-particle gages that used micrometer adjustments of the source–detector spacing to increase the measurement range on thin film thicknesses. Davison[4] and deCroes et al.[5] developed thin-film gages that used a variable ambient pressure to increase the measurement range. Hallowes and Hodgson[6] developed a gage to measure relative humidity by measuring the density of a hygroscopic film placed between the source and detector. Teranishi[7] did developmental work on a gage for the measurement of thicknesses of thin polymers.

Thick Sources. Thick (distributed energy) sources can also be used and have a range of measurement equivalent to the total range of the alpha particles minus the range in the detector window. The sensitivity is quite satisfactory, although not as high as that for thin sources on the basis of equivalent source intensities. However, the advantage of a much wider range of measurement without adjustments in source–detector spacing (and attendant calibrations) or ambient pressure makes this more attractive for most gaging applications. Such sources likewise can be used for the measurement of film thickness or gas properties.

Bretler and Steiger[8] used thick alpha-particle sources with radiographic film to measure the distribution of mass in thin films. Schumacher[9] discussed the use of thick sources and detectors with variable window thickness to measure film thickness and the properties of gases. Gardner and Ross[10,11] developed and demonstrated a gage of this type for measurements on gas pressure and further developed a continuous gage for the measurement of gas density, gas pressure, and thickness of thin films.

Gages of this type have been considered by Gilly[12] for use as altimeters and in the stabilization of the atmosphere inside spacecraft; by Howard et al.[13] for application on high-altitude constant-level balloon flights; and by Eggelsmann and Rasp[14] to measure the area of absorbers placed between the source and the detector. Gerrard[15] has summarized the potential use of both thin and thick alpha-particle sources in densitometers for measuring the density of the Martian atmosphere. Another alpha densitometer is described by Brandt et al.,[16] in which the response can be tailored to given specifications by mounting a suitably contoured baffle on the alpha source and thereby changing the energy distribution of the source.

Residual Energy. The residual-energy transmission principle involves detector systems that give a response proportional to the energy of the alpha particles reaching the detector. An ion chamber operated in the mean level mode achieves this. Both thin and thick sources may be used, and measurements can be made of film thicknesses or gas properties.

In applying this principle it is useful to have knowledge of parameters (e.g., dE/dx, range–energy relations) related to the stopping of alpha particles. A few experiments aimed at determining these parameters are reviewed first.

Ellis et al.[17,18] described a way of measuring the relative stopping powers of solid or liquid thin films by comparison with those of gases. The method involves determining the change in range of alpha particles with and without the thin film in a gaseous medium. The stopping powers of polystyrene film relative to that of acetylene gas were reported. Ellis et al.[19] further extended this measurement to relative stopping power between water and water vapor. No anomaly in the mass stopping power was detected. Gobelli[20] used thin foils and a CsI(Tl) scintillation crystal to determine range–energy relations for Ge, Si, InSb, Al, Cu, Ag, and Au.

Kerr et al.[21] measured the molecular stopping cross section of gases as a function of alpha energy for air, N_2 Ar, Kr, CO_2, and CH_4 for an energy range of ~0.5 to ~5.5 MeV. The measurements were made very precisely using a thin ^{241}Am source and a surface-barrier detector.

Early use of the residual-energy principle was made by Shirodkar[22] in measuring the temperature of a coal-gas flame, based on the assumption that the alpha stopping power in the flame is proportional to the temperature of the flame. Schopper et al.[23] extended this to measure temperature changes at atmospheric pressure. Shaw[24] described an alpha gage to determine film thickness in the range 0 to 25 g/m^2 using a radium source and an ion chamber. Chester and Newman[25] measured aqueous–organic liquid interfacial area by mixing an alpha emitter in the aqueous phase and using a liquid scintillator for the organic phase. Perette and Maugest[26] suggested the possibility of using alpha particles for on-line industrial measurement of the surface weight of thin materials by using a two-path differential method to compensate for variations in the absorbing medium between the source and the detector. Ballinger[27] described an automatic frost-point hygrometer for measuring the distribution of water vapor in the stratosphere; the frost deposit is sensed through the energy attenuation of alpha particles. A pressure gage for a wide range of pressures was discussed by Khavkin.[28] It uses two alpha sources and two ion chambers in one container.

A mathematical model to calculate the residual-energy spectrum of alpha particles, emitted from a circular alpha source coaxial with a circular detector and surrounded by a gas of known stopping power, was discussed by Cehn.[29] He also experimentally investigated the possibility of using this model for the quantitative determination of the composition of binary gas mixtures. An extension of this model is discussed by Gardner et al.[30]

Energy Shift. Application of the energy-shift transmission principle utilizes detector systems capable of alpha-particle spectrometry in conjunction with sources that are essentially monoenergetic. If the response of this system is taken as the average energy of the alpha-particle spectrum, then it can be related to film thickness or to the various properties of gaseous samples. This particular application requires considerably more sophisticated instrumentation as well as manipulation of the raw data to obtain the desired response.

Anderson[31] described a thickness gage in which the shift in average energy of alpha particles is measured, using a silicon detector and a pulse-height analyzer. Accuracy better than 1% could be obtained for Mylar foil thickness of the order of 1 mg/cm^2. Benveniste et al.[32] used a similar device to obtain relatively fast topographical mappings of thin foils. Nonuniformity in thickness of the order of 5 μg/cm^2 could be detected.

Another thickness gage capable of measurements in the range 5 mg/cm^2 to 0.5 μg/cm^2 was reported by Farouk et al.[33] Energy loss through the foil was compared to equivalent thickness of air. Reported accuracy of this gage was better than 10%. Lewis[34] described an alpha-particle thickness gage capable of measuring thicknesses up to 12 mg/cm^2 using 8-MeV alpha particles and a simple empirical formula derived from existing dE/dx data. Gilly and Jourdan[35] tested an altimeter for stratospheric balloon flights using the energy-shift principle and found the results to be identical with those given by a barometer.

Elastic Scattering

By collimating a beam of alpha particles into a sample and collimating a detector capable of spectrometry, it becomes possible to detect those alpha particles scattered within a small differential solid angle about a fixed angle from the incident beam. When a monoenergetic source and thin samples are employed, a spectrum is obtained with peaks at discrete energies that correspond to the various elements present in the sample. The lowest energies correspond to elements of lowest atomic number, since the largest energy losses occur for elastic-scattering events involving target nuclei of about the same size as the alpha particle. The scattered energies are related directly to the target-nucleus mass and the scattering angle. Both the qualitative and quantitative elemental analyses of thin solid or liquid samples are possible, since the scattered energy is related to atomic number and the intensity at that energy is proportional to the amount of element present. (Note that thick samples give rise to stepped changes in energies as opposed to discrete energy peaks for thin samples.) This technique is most sensitive for low-atomic-number elements (say, less than about 20), since the energy loss through scattering from high-atomic-number elements begins to approach a constant value as the atomic number increases.

The main use of this principle has been in chemical analysis of surfaces and thin films. Turkevich[36] described the use of large-angle scattering of heavy charged particles for this purpose. A ^{244}Cm source and surface-barrier detector were used in this study. A good description of this technique is given by Semmler et al.[37] Using a 1-mCi source, 4-cm^2 sample area, and counting times of the order of 1000 min, the minimum detectable density thicknesses for most elements is from about 2 to 10 μg/cm^2. Patter-

son et al.[38] extended this principle to the possibility of remote light-element analysis in the space program. Elements ranging from Be to Ca were determined on surfaces. Sensitivity was especially high for carbon and oxygen. Several papers[39-49] dealing with the application of this technique to Surveyor lunar landing probes and the findings from lunar surface chemical composition have been published since then.

Ziegler and Baglin[50] employed alpha scattering to determine the profile of arsenic impurity in 2 μ of substrate in silicon. The profiles measured were shown to be as accurate as those obtained by neutron activation, by differential Hall measurements, or by differential resistivity measurements. Burke and Tippett[51] reported the use of Rutherford scattering to determine foil thicknesses of high-Z elements.

Interaction Effect

Alpha-particle interaction effects in themselves are also useful for gaging. These include the ionization of gases, the excitation of characteristic X rays, and the variable collimation of alpha-particle beams by ancillary mechanical sensors so that analog signals can be digitized.

Ionization of Gases. If a gas is ionized by radiation, the resulting current (measured in the gas by an ion chamber operating in the mean-level mode) can be uniquely related (1) to pressure and temperature for a gas of fixed composition; (2) to composition for a binary mixture at constant temperature and pressure; and (3) to the mean linear flow rate for a flowing gas of fixed composition at constant temperature and pressure. The primary advantage of alpha particles in these gaging applications is that the ionization density is much higher than that for other radiations and the source intensity required for a given current is reduced accordingly.

As early as 1946, Downing and Mellen[52] reported the use of ionization caused by alpha particles for measuring pressure in the range below 10 torrs by replacing the filament of a conventional ion gage with a radium source. The range of measurement is greater than with a conventional ion gage because there is no danger of filament burnout at the higher gas pressure. A very similar ion gage for latent heat measurement was described by Beynon and Nicholson.[53]

Various other applications of the ionization principle have appeared since then. Polonnik[54] used alpha particles to neutralize the static electricity generated while rolling ribbons of synthetic fibers. Zhdanov and Makarov[55] invented a fire-alarm unit based on the decrease in ionization current as smoke particles enter an ion chamber containing an alpha source. Kobayashi[56] developed an accurate method for detecting melting point by placing a small amount of ice crystals on a RaD-E-F source inside an ion chamber and noting the temperature at which the ionization current dropped abruptly as the crystals started to melt and spread over the source. Kamienski et al.[57] described a ^{239}Pu electrode for ionizing a liquid medium and determining the electric potential across its interface with another liquid medium. They also applied this method to liquid–air interfaces.[58] Berio[59] reported on using eight ^{241}Am sources on a lightning rod to promote the ionization of air. The effective radius of the rod was 250 m.

The ionization principle has been successfully applied to flow measurements in gases. Mann[60] reported such measurements in air flowing at Mach 2. In this application, a small volume of air was ionized periodically, and the time of flight of the ionized packet was measured between two fixed reference points downstream from the source. The ions were detected by induction without the use of probes. Measurement accuracy reported was 2%. Hribar[61] discussed a modified continuous-ionization method for gas flow rate measurement at low flow speeds. He also considered the effects of gas composition, pressure, and temperature on the measurement.

Excitation of Characteristic X Rays. Alpha particles can be used to excite the characteristic X rays of elements in solid, liquid, or gaseous samples. If X-ray spectra with a suitable detector system are measured concurrently with this excitation, both qualitative and quantitative elemental analyses of the samples can be made. Alpha particles are advantageous for measurements of very thin surfaces or for very-low-energy characteristic X rays (where other sources of radiation give rise to low-energy background radiation in the energy range of interest).

Friedman[62] demonstrated that characteristic monoenergetic X rays can be produced efficiently with alpha sources. Sellers and Ziegler[63] and Sellers et al.[64] reported the feasibility and added safety of millicurie alpha sources for X-ray analysis. They also compared the results from alpha excitation with beta, proton, and triton excitation. An extensive review of their work prior to 1967 is given in a later article by Sellers et al.[65] Campbell et al.[66] also have covered this same area of research in their elaborate review of X-ray absorption and emission. A similar review of alpha excitation of X rays is given in an article by Fuccillo[67]

on radioisotope instruments in industry and geophysics and by Martinelli et al.[68,69] on X-ray analysis by α, β, X, and γ sources.

Various papers dealing with special applications of this technique have appeared in the literature. Plaksin et al.[70] reported using an alpha source for elemental analysis of ores and beneficiation products. Putman[71] used this technique to measure the ash content of burned coal as a means of enhancing the full calorific value of the fuel. Uchida et al.[72] developed an alpha X-ray analyzer for light element analysis for controlling the mixing of raw materials in cement. An X-ray analyzer using 35-MeV alpha particles for oxygen analysis in aluminum was reported by Deyris and Albert.[73] McKinney[74] described a French X-ray fluorescence analyzer for light elements using alpha excitation. Adler and Trombka[75] discussed the feasibility of using an alpha-excited X-ray analyzer for rock analysis on the lunar surface.

Digitizing Mechanical Sensors. In certain applications, such as in process control with on-line computers or remote sensing and subsequent telemetering of responses, it is desirable to have ordinary measurement responses in digital form. When mechanical sensors are involved which are capable of moving small diaphragms or shutters, a small alpha-particle source and detector can be placed on either side of the diaphragm to give a digital response that is representative of the response of the mechanical sensor. Applications include Bourdon pressure gages and D'Arsonval voltmeters.

Hyde[76,77] described a digital method for measurement of process variables based on modulation of the radiation from an alpha source. It was designed for use in conjunction with transducers for pressure and temperature measurements. The digital readout was obtained with a pulse counter. The principal advantages of this method are excellent long-term stability and reasonable cost. (PSB)

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A STUDY OF PARTICULATE HETEROGENEITY EFFECTS IN RADIOISOTOPE X-RAY SPECTROMETRY*

The effects of particle-size heterogeneity on X-ray fluorescence, backscattering, and transmission in granular materials and slurries were studied at Texas Nuclear Corp. under a Division of Isotopes Development contract. Hopefully, the results could be applied in the evaluation and possible development of nondestructive radiometric techniques for minimizing particle-size effects in nondispersive radioisotopic X-ray fluorescence analysis and for particle-size determination per se.

Existing theories were reviewed, and a new theory was developed to quantitatively explain the effects caused by changing the size in mixtures of particles of equal size in terms of the physical parameters of the material and the radiation energy characteristics. The model was then extended to a more general situation of different-size-particle mixtures. Experiments on the validity of the new model showed it to be generally more consistent than existing theories and to be useful in examining a wider range of sample parameters—element concentration, packing density, and slurry dilution—and incident X-ray energy than could be done by experiment or with available sources and instrumentation.

Study of the mutual influences of particle size, chemical composition, and (slurry) solids content indicated that size determination in mixtures of particles of equal size is independent of chemical composition and is feasible through appropriate choice of X-ray energies. For different-size-particle mixtures, however, the practicability of these techniques is questionable. In both artificial and natural materials there was considerable ambiguity in the intensity–particle-size functions, and a simple radiometric technique probably cannot be devised for particle-size compensation and measurement. The need for more complex methods using combined radiometric analysis and physical sample treatment is indicated. (MG)

*Summary of USAEC Report ORO-3847-2 by P. F. Berry, Nuclear-Chicago Corp., Texas Nuclear Corp. Division, June 3, 1971.

DETERMINATION OF PHOTOGRAPHIC FILM-EMULSION THICKNESS BY NEUTRON ACTIVATION*

Supplementary Keywords: *source, neutron; measurement, thickness; activation, neutron;* ^{110}Ag; ^{239}Pu.

The thickness of the emulsion on a photographic film was determined by activation of 1 cm^2 of the film with a ^{239}Pu–Be source. A flux of 1×10^8 n/sec is required to form ^{110}Ag from the silver in the emulsion, the activity of which was measured with a 256-channel pulse analyzer. The silver content of the emulsion was of the order of 10^{-4} g/cm^2. (MG)

*Summary of article (in Russian) by I. A. Abrams, M. R. Bire, V. I. Breslav, L. L. Pelekis, and I. Ya. Taure, *Izvestiya Akademii Nauk Latviiskoi SSR*, **1970**(4): 31-36.

^{35}S BETA RADIOGRAPHY OF PAPER*

Watermarks and other structures in paper may be demonstrated by placing a beta-radioactive source—thin sheet impregnated with ^{35}S—on one side of the paper and a photographic film on the other with a source activity of 2.5 μCi/cm^2; good prints are obtained with a 15-min exposure. Flat sources impregnated with ^{14}C (0.7 μCi/cm^2) may also be used.† (MG)

*Summary of W. J. van Ooij, Beta Radiography of Paper Using a Flat Beta-Ray Source (in Dutch), *Papierwereld*, **24**(3): 67-71, 73 (1969).

†Similar British work with ^{14}C was described in *Isotopes and Radiation Technology*, **6**: 287-288 (1969).

Section

III

Isotopes and Radiation Technology

Food Irradiation

Radurization of Scampi, Shrimp, and Cod

By G. Hannesson and B. Dagbjartsson*

(Summarized by F. E. McKinney)

Supplementary Keywords: *food preservation; industry, food; seafood; source, gamma; food evaluation, subjective; food evaluation, objective;* ^{60}Co.

Abstract: *Research on the radiation pasteurization of Norway lobster tails (scampi), deep-sea shrimp, and whole cod and cod fillets is reported. At 0 to 1°C and a 200-krad dose, the shelf life of cod was extended by a factor of 3 (up to 19 days for trawl cod and 24 days for line cod). At 0 to 1°C and a dose of 100 to 200 krads, shelf life of blanched shrimp and scampi could be increased to about 6 weeks—about twice that of unirradiated specimens. This article is a summary of an IAEA publication, STI/DOC/10/124. The conclusions in the three sections of this 93-page report are supported by data presented in 29 figures and 28 tables.*

In 1968, through an agreement with the International Atomic Energy Agency, the USAEC loaned to Iceland a 35-kCi ^{60}Co irradiator that had previously been used on board the M. V. *Delaware*. Research was conducted with the irradiator to determine whether or not radiation preservation is practical under conditions that prevail in the Iceland fishing industry. The domestic market in Iceland is small, and a large amount of seafood is exported—mostly salted, dried, or frozen. About 30 thousand to 40 thousand tons of iced fish are shipped annually to the United Kingdom. The investigation concentrated on cod, scampi (Norway lobster tails), and shrimp.

RADURIZATION OF NORWAY LOBSTER TAILS

Lobster fishing, a new branch of the Icelandic fishing industry, is localized on the southwest coast, where small boats with specially designed light trawls are used. When caught, the lobster's head and carapace, with claws and legs attached, are twisted off and discarded; the unshelled tails are iced until landed. As soon as possible, the tails are washed, graded, wrapped, packed in 5-lb boxes, and frozen. Prime tails are sold for about $3.55 per kilogram after freezing, and the mean annual catch for 1960 to 1966 was 3000 metric tons. Lobster tails cannot be exported iced because the shelf life is limited by black color formation (melanosis), rapid ammonia and trimethylamine production, and microbial spoilage.

Lobsters obtained from commercial processing plants (after processing but before freezing) varied in quality, being 1 to 3 days out of the sea. Lobsters had to be processed in cans because the spines tore plastic bags. Radiation doses were 100, 200, and 300 krads; blanching times were 1 and 2 min; and storage temperatures were 0 to 1° and 5 to 6°C. Subjective evaluation was based on panel-scoring of samples for odor and flavor; objective evaluation was based on determination of volatile acid number and total bacterial count. The limits for shelf life were set as a total bacterial count of 1×10^6 per gram and a sensory score of 2 on a 5-point scale.

Results of Experiments

Irradiation alone does not prevent formation of black spots. Scampi at 100 krads was lighter than scampi at 300 krads, but both were darker than controls. A chemical dip (0.45% citric acid and 0.05% ascorbic acid) was tested, but the irradiated product

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was unacceptable because of black color only 1 day later. Blanching consisted of dipping the lobster tails in boiling water for a set time—0.5 min, 1 min, or 2 min. Only the last two time intervals produced control of melanosis with irradiation. Blanching causes the shell to become lighter in color and the meat to appear cooked.

The two temperatures chosen for storage tests are the conventional storage temperatures for seafoods. Shipping tests carried out between Iceland and Vienna to evaluate procedures showed that, even when surface-shipped and re-iced only once en route, the 300-krad lobster tails were of fair quality. Shipment by air of samples given 100 krads and blanched for 2 min provided a product about equal to a frozen reference product.

The bactericidal efficacy of blanching alone, radiation alone, and blanching and radiation together was tested. Blanching for 0.5, 1, or 2 min reduced the bacterial number by 1.2, 2.0, or 2.5 log cycles, respectively. Doses of 100, 200, and 300 krads reduced the bacterial number by 2.2, 3.3, and 3.7 log cycles, respectively. The combination of blanching and irradiation reduced the bacterial load by 4 to 6 log cycles, which resulted in less than 100 microorganisms per gram for all treatments.

As might be expected, the 300-krad dose prolonged shelf life the longest of the three doses. Blanching was very effective if applied for 2 min, but blanching for 1 min gave marginal shelf-life extension. The combined effect was synergistic in all cases.

Discussion of Results

The variation in raw material from batch to batch caused difficulty with statistics but also more closely simulated the situation that might occur in a commercial operation. The radiation dose used must result in the greatest extension of shelf life with the least change in the product. Above 300 krads, undesirable changes occurred in lobster, and the expert sensory-evaluation panel could easily distinguish the 300-krad sample. The limit for sensory evaluation was usually reached at a later date than was the microbial limit of 10^6 bacteria per gram.

Among the few objective evaluation tests for seafood are tests for trimethylamine (TMA), total volatile bases (TVB), and volatile acid number (VAN). The VAN test was used because it seemed most likely to give indicative results and because it is the simplest of the three tests to perform. However, it gave uneven results for irradiated and blanched lobster. Correlation was possible in lobster blanched less than 1 min, and the VAN test seemed indicative of spoilage for raw material and untreated lobster.

The bacteriological tests involved culture and incubation followed by count of the plate. The lobster lives in an environment that seldom exceeds 10°C, and consequently the microflora is predominantly psychrophilic. Pathogenic organisms are introduced by contamination during processing with the exception of *Clostridia* species. In this study, microorganisms were not identified. Because the raw material was received from commercial sources, the initial number of bacteria varied considerably—up to 10^6 per gram. Radiation was used for pasteurization purposes; the bactericidal effect of blanching was inherent but incidental to the process. For microbiological control, the combination of 2-min blanching and 200 to 300 krads was most effective.

Storage at 0 to 1°C in general gave twice the shelf life of storage at 5 to 6°C regardless of other factors. A combined blanching–irradiation treatment gave a storage life of 2 to 3 weeks at the higher temperature, while untreated lobsters could be stored at 5 to 6°C for only 1 to 2 days. Untreated lobsters lasted 5 to 6 days at 0 to 1°C, but lobsters irradiated after a 2-min blanch could be stored about 6 weeks.

Additional Information

Lobster fishing is seasonal, and, because many of the same plants also process whitefish, the peak catch of which occurs at a different time, there is a basis for a year-round commercial irradiation plant. Such a plant would be expected to process about 6000 metric tons annually, of which 1000 tons would be lobster that would have to be processed within 3 months. The economic implications are difficult to assess, and speculation is futile until consumer acceptance is established and food laws have approved radurized seafoods.

Conclusions on Irradiated Lobster

The sensory evaluations and the total bacterial count were in close agreement, but VAN determination was not reliable for the irradiated and blanched product. Irradiation alone extended shelf life, but blanching was also necessary to inhibit melanosis. The most promising results were obtained when lobster was blanched for 2 min and irradiated to 200 and 300 krads, with storage at 0 to 1°C. Further research is needed on the effect of raw-material quality on storage life, identification of the microflora, the feasibility of

blanching at sea, the economics of using radiation in the Icelandic fishing industry, and consumer acceptance of an irradiated product.

RADURIZATION OF DEEP-SEA SHRIMP

Deep-sea shrimp or prawn, *Pandalus borealis*, are of economic significance worldwide. Shrimp of the species caught off Iceland are small (1 to 2 g of meat) and high priced, with an annual catch of about 2000 metric tons, primarily from October through April. The shrimp are stored on ice until processed on shore, where peeling, precooking, freezing, and packaging take place. The same methods were used in this study as in the lobster study, except that the shrimp were packaged in polyethylene bags and TMA was determined rather than VAN. The experiments were divided into five groups: effect of treatment before irradiation (blanching, low-dose preirradiation, and storage at 0 to 1°C); effect of blanching followed by peeling and storage; comparison of combined treatments (double irradiation, blanching plus irradiation); storage after blanching for days before further treatment; and irradiation of commercially prepared shrimp.

Results of Experiments

Blanching for 2 and 5 min reduced the log bacterial number by 3.89 and 4.03, respectively. In one trial a preirradiation dose of 60 krads reduced the bacterial number by 0.59 log cycle, and an additional reduction of 1.12 log cycles was caused by a 200-krad dose 2 days later. Blanching for 2 min, followed by peeling and irradiation at 100 krads, reduced the log bacterial number by 4.92 and 2.83 in two trials. At a dose of 200 krads, the reduction was 3.10. The efficacy of delaying irradiation after blanching was difficult to assess, but, in all cases, the number of surviving bacteria was less than 100 per gram after irradiation and before determination of storage life.

Untreated shrimp can be stored 5 to 6 days at 0 to 1°C, with spoilage indicated by a green to black discoloration, which a pretreatment dose of 60 krads failed to prevent. Blanching is commonly used in the industry to facilitate peeling and prevent discoloration. Blanching for 2 min alone will increase the storage life to 10 days, although some discoloration is evident after 7 days. Increasing the blanch time to 5 min gave little additional benefit. Blanching followed by peeling extended the storage life to 12 days. The trial with a 60-krad dose followed by a 200-krad dose 2 days later produced an organoleptically unacceptable product. The longest storage life was obtained by a combination of a 5-min blanch and a 100-krad dose—about 45 days. Unpeeled, blanched shrimp were stored for 2, 5, or 7 days before irradiation to test this effect on the process; only the sample stored 7 days before irradiation showed a decrease in shelf life.

Additional Information

Raw material varied somewhat in quality, but use of the same raw material as industry uses would seem logical in dealing with practical aspects of radiation application. The main shrimping grounds are off the northwest coast of Iceland, where 10 small processing plants are now located. If a food-irradiation center is located on the southwest coast, where the main whitefish and lobster plants are, some pretreatment of the shrimp will be necessary to provide additional transport time. Blanching evidently would be sufficient so long as the trip does not take more than 5 days. Blanching has a greater bactericidal effect in shrimp than in lobsters because the shrimp are treated whole.

When the limit of bacteriological acceptability was set at 10^6 microorganisms per gram, the time of sensory rejection occurred as much as 20 days after the bacteriological limit was reached, but raising the limit to 10^7 per gram lessened this discrepancy. The sensory evaluations showed that shrimp were more sensitive to dose than were lobsters. However, lobsters were served boiled with a minimum of water, and the shrimp were steamed. Panel members sometimes preferred irradiated shrimp to control shrimp because the polyethylene packaging imparted an off-flavor to controls. The TMA number failed to correlate reliably with the sensory data.

Conclusions for Irradiated Deep-Sea Shrimp

Blanching is necessary to delay spoilage before irradiation and to prevent discoloration. Doses above 200 krads caused unacceptable organoleptic changes, but this level gave superior microbiological control. Blanched, unpeeled shrimp can be kept on ice for 5 days before irradiation without a decrease in quality; this time interval will suffice for a trip from the shrimping grounds to the food-irradiation plant. The optimum combination of treatments—5 min blanching and a 100-krad dose—allowed storage for up to 7 weeks at 0 to 1°C. If sensory evaluation is used as the standard for other criterions of end of storage life, neither TMA production nor total bacterial count was a good indicator of spoilage for the irradiated product.

RADURIZATION OF WHOLE COD AND COD FILLETS

Whitefish are the most important part of the Icelandic seafood industry, with 350 to 400 thousand metric tons being landed annually, of which 200 to 250 thousand metric tons are exported as prime quality frozen fillets and about 20 to 40 thousand metric tons are exported as iced, whole fish. Of the whitefish species caught, cod is by far the most abundant, although haddock generally draws a higher price on the market. Special consideration was given in this study to methods of catching the fish because the different methods give different quality fish—inshore fishing with hook and line producing the highest quality and net fishing an inferior quality but a much greater quantity. Trawl fishing in general produces better quality than net fishing but, because boats stay out longer, may result in an inferior-quality fish.

The raw material for studies with fillets was obtained from commercial plants after filleting but before packaging and freezing. Whole, gutted cod were obtained directly from fishing vessels. Dosimetry, packaging, storage, and analytical methods were similar to those used in the shrimp and lobster research. Irradiation doses tested were 50, 100, 200, and 300 krads; fillets irradiated at the highest dose showed distinctly lower organoleptic scores. A dose of 200 krads was optimum for fillets, while 50 krads was used on whole cod, which in some experiments were filleted and given a second dose of 150 krads.

A different sensory evaluation was used with cod, based on the odor, which entailed retraining the test panel; instead of a low score being the limit, the time of rejection by the panel was used.

Results of Experiments

Unirradiated, line-caught cod fillets stored at 0 to 1°C lasted 7 days; net-caught cod and trawl-caught cod lasted only 5 days. Storage at 5 to 6°C reduced shelf life to 3 to 4 days for all fillets. Irradiation to 200 krads increased the life of line cod at 0 to 1°C to 24 days; net cod lasted 20 days and trawl cod 19 days. Irradiated fillets stored at 5 to 6°C lasted 13 days for line cod and 11 days for net and trawl cod.

Whole line cod stored at 0 to 1°C were rejected after 12 days; whole net cod were rejected in only 6 days. Irradiation increased storage times to 21 and 13 days, respectively. Net-caught cod used in this determination were markedly lower in quality than the line-caught cod. Fillets (from whole line cod that had been given a 50-krad dose and stored for 3, 7, or 12 days before filleting) re-irradiated to 150 krads could be stored an additional 18 to 20 days. Cod filleted in the laboratory, where extra care was taken, showed an increase of 9 days in storage time over commercially filleted cod.

The VAN determined for unirradiated cod and whole cod given 50 krads could be correlated with time of sensory rejection, but correlation at higher doses was unreliable. The TMA values rose shortly before rejection time in irradiated cod fillets, and some correlation was possible between TMA and sensory evaluation. Correlation was best in fillets cut from irradiated whole cod.

The initial bacterial log number in all fillets averaged 5.9. Line cod given 100 krads showed a 1.75 log number reduction, with a 2.45 log number reduction after 200 krads. Net cod showed a 2.9 log number reduction after a 200-krad dose. Trawl cod showed a 2.4 log number reduction after 100 krads and 3.3 log number reduction after 200 krads. The rejection level in the bacterial studies was set at 10^7 microorganisms per gram. At 0 to 1°C, a 200-krad dose prolonged storage by a factor of approximately 2 as compared to unirradiated storage. Irradiated fillets of line cod had the longest storage life, about 2 days longer than net cod and 3 days longer than trawl cod. For unirradiated cod, the limit of 10^7 per gram was reached at about the same time as the other two assessment limits, but for irradiated cod fillets, this limit was reached well before the other limits. However, in experiments with whole cod, the limit of bacteriological assessment was reached later than the limits of the other assessment methods.

Additional Information

The aim of this study was to investigate the effects of initial quality on the quality of the irradiated product and on the extension in shelf life. Both are known to affect quality of iced and frozen cod. The difference in quality between line-caught cod and net-caught cod is the result of the catching method; the quality of trawl-caught cod is a function of how long the product has been iced before processing. The odor-evaluation test produced satisfactory results. Of the chemical tests, determination of VAN was less useful than determination of TMA. The bacteriological limit of acceptance was reached sooner in irradiated fillets than was the organoleptic time of rejection. A dose level of 100 krads was judged to be inadequate to obtain the necessary storage-life extension for irradiated fish to be shipped iced to foreign markets.

Irradiation followed by storage and reirradiation was tested, but fishing vessels land their catch daily (except for trawlers), which makes shipboard irradiation facilities of doubtful benefit.

Conclusions on Radurized Cod

The odor test developed for sensory evaluation was easy to carry out and gave reliable results. The TMA determination correlated fairly well with the odor test, but the VAN determination showed poor correlation at doses above 100 krads. Irradiated cod fillets reached the limit of bacterial acceptance, 10^7 microorganisms per gram, before the rejection time based on organoleptic acceptance was reached; however, bacterial limits on whole cod were reached later than organoleptic limits. Commercially procured line cod fillets could be stored 24 days at 0 to 1°C after a 200-krad dose. Commercial operation of a shore irradiation plant is considered feasible, with filleting of cod by machine recommended to minimize bacterial contamination.

Irradiation of Fruits and Vegetables—1966 to 1970

By P. Markakis and R. C. Nicholas*

(Summarized by F. E. McKinney)

Supplementary Keywords: *food preservation; industry, food; source, gamma; ^{60}Co.*

Abstract: *Results are summarized of research on radiation preservation of a number of foods, conducted by Michigan State University from April 1966 to December 1970. Products studied include asparagus, blueberries, strawberries, sweet cherries, peaches, sweet corn, apple juice, and mushrooms. This article is a summary of USAEC Report COO-1592-37. A bibliography of publications resulting from this research is included.*

Under a contract with the Division of Isotopes Development, USAEC, from April 1966 to December 1970, researchers at Michigan State University investigated the possibility of extending the marketing period of fresh fruits and vegetables. Irradiation extends market life in two ways—reduction of microbial load and delay of ripening. The beneficial action of radiation is often supplemented by other techniques, such as heating, refrigerating, and using chemical dips or controlled atmospheres. The source of radiation used in these studies was the 50-kCi ^{60}Co source at Michigan State, except in experiments with mushrooms when the 8-kCi ^{60}Co source at the Phoenix Memorial Laboratory, Ann Arbor, was also used. Research is summarized separately for each food product.

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ASPARAGUS

Spears were given doses of 10 to 100 krads and stored at 4 to 10°C. Irradiation slightly retarded spear lengthening but had little or no effect on water loss, color, texture, or fiber-plus-ash content. There seems to be no advantage derived from irradiation of this product.

BLUEBERRIES

Doses of 150 and 300 krads significantly reduced the freeze-drying time (Table 1) of blueberries.

Table 1 Weight Loss of Jersey Variety Blueberries During Freeze Drying

Drying time, hr	Dose, krads	Weight loss, %
0	0	0
	300	0
3	0	10.3
	300	14.1
8	0	33.7
	300	41.8
18	0	61.8
	300	73.8
30	0	71.8
	300	83.1

Radiation-induced softening of the fruit could be offset by dipping the berries in 0.5% Ca^{2+} solution (from $CaCl_2 \cdot 2H_2O$) for 1 hr before irradiation.

STRAWBERRIES

Microbial reduction was attempted with doses of up to 800 krads. The total count decreased more rapidly with dose than did the mold-plus-yeast count, which indicated that the bacteria were more radiation sensitive than the molds and yeasts. A 400-krad dose gave a 99% reduction of the total microbial load. Packaging trials showed that overwrapping the strawberries in pint wooden boxes with plastic film did not decrease the number of lesions or maintain firm texture. Treatment with the calcium dip did not show consistent results, although irradiation and soaking gave better spoilage control than soaking alone.

SWEET CHERRIES

Doses of 100 to 800 krads reduced microbial growth, but irradiated cherries were softer than controls. Soaking, either before or after irradiation, in the 0.5% Ca^{2+} solution for 1 hr did control softening. In any case, cherries with stems attached kept better than those without stems. No advantage was shown in a combination treatment where the cherries were dipped in water at 46 or 50°C for 4 min and irradiated with 100 or 200 krads.

PEACHES

Red Haven peaches were treated with hot-water dips (46 and 50°C) and radiation doses of 50 or 100 krads, but shelf life was not extended by any treatment, separately or in combination. Heating caused a mottled appearance, and irradiation caused softening.

SWEET CORN

The quality of corn in storage is related to the moisture content, which indicates the amount of conversion of sugar to starch. An attempt was made with doses of 30 and 300 krads to improve moisture retention by irradiation. This objective was not achieved, although the storage period tested was only 14 days.

APPLE JUICE

The possibility of pasteurizing apple juice by radiation was explored. A dose of 300 krads reduced total and yeast-plus-mold counts to 1%. Irradiation lightened the color of the juice but had no effect on its viscosity. Heat treatment at 49°C for 30 min, either before or after irradiation, had a good effect. The juice could be stored an additional 15 to 20 days at 3 to 4°C, although the combination treatment further lightened the color of the juice. No effect on flavor was noticed.

MUSHROOMS*

Of all the commodities tested in this work, fresh white mushrooms, a highly perishable product that can be stored only 1 day at 50°F (10°C) or 5 days at 32°F (0°C), responded best to irradiation as a means of prolonging shelf life. The undesirable changes that occur during storage—opening of the cap, darkening of the gills, elongation of the stem, darkening of the cap and stem, and shriveling—were substantially retarded by doses of only 25 to 100 krads. One problem encountered during storage of mushrooms is to provide sufficient oxygen exchange to prevent "suffocation" while avoiding excessive dehydration, which promotes browning. One-quart polyethylene tubs were found to be excellent for storage.

Measurements

Two indexes of quality change were derived for mushrooms—degree of opening and opening ratio. Degree of opening (D.O.) is a fractional measure of the separation of the veil from the stem and is carried out by slipping a transparent disk, marked every 36°, down over the stem. When the veil is completely closed, the D.O. is 0, and when completely separated, the D.O. is 1. The measurement is made easily and is nondestructive, but the variability is large and correlation with other changes is low. The opening ratio (O.R.) is defined as $O.R. = (b + c)/[a - (b + c)]$, where a is the diameter of the cap, b is the distance from one gill tip to the stem, and c is the distance from the opposite gill tip to the stem. The $(b + c)$ is the total opening gap, less the stem diameter, and $[a - (b + c)]$ is an "effective" diameter. The measurement is destructive, but it

*See also the summary of USAEC Report COO-1592-36 in *Isotopes and Radiation Technology*, 9(3): 303-305 (Spring 1972).

has the advantage of combining measurements of expanding cap and shrinking stem in one index.

Mushroom height, shear resistance, reflectance, and respiration were also measured during these experiments.

Results

On the basis of cap opening, the shelf life of mushrooms was extended 3- to 5-fold by irradiation (Table 2) in the dose range 25 to 100 krads, with subsequent storage at 5°C. There was wide variation from test to test, presumably caused by such variables as the particular "break" harvested, maturity at harvest, and moisture loss during storage. The rate of change of O.R. was nearly linear up to O.R. = 1.

Table 2 Changes in Opening Ratio of Stored Mushrooms

Storage		Opening ratio*			
Temp., °C	Time, days	0 krads	50 krads	100 krads	200 krads
5	21	1.00	0.55	0.60	0.50
10	15	1.95	0.60	0.40	0.40
15	4	1.20	0.55	0.50	0.50

*O.R. = $(b + c)/[a - (b + c)]$. See text.

Changes in dimensions are unequally influenced by irradiation (Table 3). Irradiation reduced the rate of cap opening, but no dose effect was apparent. Mushroom height changed in a manner inconsistent with total dose, as did weight loss. Stem diameter decreased for all treatments. The color of irradiated mushrooms was lighter than that of controls, and neither taste nor texture was affected by the treatment.

In another experiment, gamma radiation was shown to be more effective than 1-MeV electrons in inhibiting cap opening, especially when mushrooms were irradiated in a stem-down position. (FEM)

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Table 3 Physical Changes in Irradiated Mushrooms

Property	Original value (average)	Storage, days	Change, %				
			0 krads	10 krads	20 krads	50 krads	100 krads
Cap dia.	1.37 in.	7	16.4	5.7	7.4	5.3	3.5
		11	25.2	7.1	7.8	4.7	2.5
Height	1.29 in.	7	31.8	39.6	47.4	34.0	9.7
		11	35.5	41.7	49.4	35.5	19.7
Stem dia.	0.64 in.	7	−27.2	−21.1	−16.6	−15.0	−16.2
		11	−31.2	−26.7	−22.6	−20.6	−21.3
Weight	12.20 g	7	−11.3	−15.8	−14.9	−12.0	−13.0
		11	−14.9	−19.7	−17.1	−14.6	−15.4
D.O.	0.0	7	1.0	0.8	0.7	0.4	0.0
		11	1.0	0.8	0.8	0.4	0.0

GAMMA-RAY TREATMENT OF TASTES AND ODORS*

In laboratory studies, the musty, earthy odor of water artificially contaminated with *Streptomyces* was decreased by treatment with 8.4×10^3 rads of ^{60}Co gamma radiation just as well as by activated carbon. Activated carbon, although the most efficient agent for removing earthy odors from water supplies, is very inefficient and expensive when the odorous substance is present in low concentration. Gamma radiation was equally efficient with high and low concentrations. A cost of 30 cents per 10^3 gal was estimated for applying 10^4 rads with ^{60}Co to a 10^6 gal/day plant. However, since the dose required is low, spent fuel elements could conceivably be used, with a decrease in cost.

*Abstract of article by Ann H. Vajdic, *J. Amer. Water Works Ass.*, 63: 459-461 (1971). Some Russian work on gamma irradiation of mineral water to destroy unpleasant odors is reported by V. G. Bregvadze in *Action of Gamma Radiation on Nonalcoholic Drinks, Wine, and Brandy*, Chap. I, pp. 4-18, Izdatel'stvo Pershchevaya Promyshlennost', Moscow, 1970 (in English: ORNL-tr-2516, machine translation edited by Martha Gerrard and P. S. Baker).

Results of Strawberry Shipping Studies

By E. C. Maxie*

(Summarized by F. E. McKinney)

Supplementary Keywords: *food preservation; industry, food; source, gamma; fruit;* ^{137}Cs.

Abstract: *Seven test shipments of irradiated strawberries were carried out during the 1970 California harvest season—two by sea to Honolulu, four by truck to Atlanta, and one by truck to Minneapolis. Dose levels were 150 and 235 krads. Irradiation was significantly effective in controlling decay in the overseas shipments, but in transcontinental truck shipments the treatment was less effective, although still better than not using irradiation. The dose of 150 krads was slightly superior to the 235-krad dose. This article is a summary of USAEC Report UCD-34P80-9.*

Strawberries have long been considered one of the most promising fruits for application of radiation to inhibit decay and extend shelf life. Research at the University of California, Davis, has been supported by the USAEC Division of Isotopes Development since 1961 to investigate this possibility. During the 1970 harvest season, shipping studies were continued to evaluate the commercial use of the irradiation technique.

*Department of Pomology, University of California, Davis. Original title: Radiation Technology in Conjunction with Postharvest Procedures as a Means of Extending the Shelf Life of Fruits and Vegetables.

HARVEST PROCEDURES

Berries were picked by hand by eight especially chosen expert pickers and immediately placed in a forced-air cooler at 35 to 37°F for transportation 20 miles to Salinas, Calif., for irradiation. The berries were returned in the same cooler to Watsonville, where trucks were loaded for transcontinental shipping or for transfer to Oakland and overseas shipping. Standard practice in the industry is to place strawberries in the middle of a van, with other produce loaded above the axles, to prevent shipping damage to the delicate strawberries.

IRRADIATION PROCEDURE

For these tests, the Portable Cesium Irradiator (PCI) was used instead of the Mobile Gamma Irradiator (MGI), which has been used for these studies in previous tests. The PCI has a stationary ^{137}Cs source; material to be irradiated is inserted above or below this source. The MGI has a ^{60}Co source, and material to be irradiated is carried through the irradiator on a conveyor. The PCI is thought to cause less mechanical injury to strawberries than does the MGI.

The two irradiation doses used were 150 and 235 krads. These were absolute maximum doses, and, because uniformity ratio was 1.35, the minimum doses received were 111 and 173 krads.

RESULTS

Table 1 summarizes the end results of the shipping studies. In all but one case, the irradiation treatment reduced the amount of decayed fruit; however, in only one test was the difference statistically significant. Irradiation at 150 krads seemed to give slightly better decay control than did 235 krads.

Table 1 Percent Marketable Shasta Strawberries After Irradiation and Shipment

Shipment method	Destination	Date shipped	Dose, krads	Percent marketable fruit		
				Before shipment	On arrival	After 3 days of simulated storage
Truck	Minneapolis	7/16/70	0	98.92	94.93	90.31
			150		91.97	91.46
			235		91.52	90.92
Truck	Atlanta	7/31/70	0	98.54	93.66	86.67
			150		95.75	93.19
			235		94.80	90.05
Truck	Atlanta	8/26/70	0	98.67	85.26	87.59
			150		88.47	86.45
			235		90.96	86.60
Truck	Atlanta	9/17/70	0	96.27	85.01	45.63
			150		91.17	81.83
			235		92.70	86.97
Truck	Atlanta	10/1/70	0	98.40	94.43	84.49
			150		95.58	92.99
			235		94.78	89.80
Surface ship	Honolulu	8/16/70	0	97.96	49.98	19.66
			150		92.64	80.34
			235		89.89	84.18
Surface ship	Honolulu	8/21/70	0	98.66	95.51	85.26
			150		97.10	87.50
			235		94.97	85.60

DISCUSSION

When research on this project began in 1961, most California strawberries were transported long distances by refrigerated railcars. In 1970, refrigerated trucks hauled 82% and airlines the other 18% of the strawberry crop; truck time to the east coast is only 3 to 4 days. Loss during train shipments averaged 25% and sometimes reached 50%; loss by truck is thought to average less than 10%. Further changes in the situation reflect greater care in harvesting and handling: (1) dramatic improvement in picking control, (2) use of forced-air cooling, (3) rapid delivery to the cooler after picking, (4) use of plastic sheets under strawberry plants to reduce field infections, (5) improved physical characteristics of the berries, and (6) improved ride and temperature control in refrigerated trucks. The extremely long time required to collect data for the petition to the Food and Drug Administration has permitted the development of additional and improved technology that has reduced strawberry loss to near acceptable levels without the use of radiation. However, two possibilities exist for future use: (1) extending the market to overseas areas, and (2) unionization of the labor force in California, which might reduce field quality and reintroduce the need for irradiation in the domestic market.

(FEM)

PRESERVATION OF BETEL LEAVES ATTEMPTED*

One of the more unusual applications of ionizing radiation has recently been reported. Betel leaf is a popular dessert in Southeast Asia and must be transported to consumers in West Pakistan by air freight from East Pakistan at high cost because of the short shelf life of 4 to 7 days. If a preservation method could be found to enable shipment of betel leaf by sea, costs would be reduced and new markets could be developed. A 200-Ci ^{60}Co source was used to provide doses ranging from 10 to 200 krads—with plant hormones, a mild heat treatment, and controlled atmospheres also tested. All attempts with radiation—alone or in combination with other treatments—failed. The only method that extended shelf life was storage in a controlled atmosphere with a high concentration of CO_2 and a low concentration of O_2.

(FEM)

*Abstract of Preservation of Betel Leaves by Irradiation, Chemical Treatment, and Controlled Atmosphere Storage, by A.T.M. Faizur Rahman and A. K. Siddiqui, Atomic Energy Centre, Dacca, East Pakistan, Report AECD/RB/11, October 1970.

NUCLEAR TECHNIQUES AND THE GREEN REVOLUTION

Addendum to the IAEA Annual Report to the Economic and Social Council of the United Nations for 1970-1971, IAEA, Vienna, 1971 (INFCIRC/146/Add. 1).

This addendum is a 31-page popular description of the ways in which nuclear techniques have contributed to the "Green Revolution"—the growing of new high-yielding cereal varieties in the developing countries. The nuclear techniques that have been used include tracing, activation analysis, and irradiation.

Irradiation has been mainly used to induce mutations, i.e., the new varieties that are producing more food. In addition to higher yields, the radiation-induced mutations must have certain nutritional value, storage and processing characteristics, and consumer-pleasing characteristics. Radiation is also being used in a massive sterile-male program to eradicate certain insect pests. Activation analysis has been used to investigate plant materials for the presence of elements necessary for nutrition. Tracers have been used to optimize fertilization; the new varieties respond best to correct amounts of fertilizer, and preventing excess fertilization prevents pollution of lakes and rivers with organic material. Tracer techniques have also been used to evaluate water supplies in proposed growing areas.

In an appendix are listed the research programs, symposiums and conferences, publications, and training courses that the IAEA has helped provide in this field. (FEM)

Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Research and Development Studies into the Basic Technology of Radiation-Induced Ionic Polymerization Including Small-Scale Pilot-Plant System*

Supplementary Keywords: *source, gamma; industry, plastics;* ^{60}Co.

Abstract: *Basic laboratory studies and engineering pilot-plant research on the radiation-induced ionic polymerization under superdry conditions are briefly described. This is a summary of USAEC Report ORO-3667-1, final report, June 1, 1967, to May 31, 1971. Original report date June 1, 1971.*

North Carolina State University (NCSU), under contract to the USAEC Division of Isotopes Development, has conducted studies on the basic technology of radiation-induced ionic polymerization. For many years the radiation polymerization of vinyl monomers was believed to occur only through free-radical formation. In 1957, however, it was discovered that isobutylene would polymerize to a high-molecular-weight polymer when irradiated at low temperatures. Since this monomer was not known to polymerize to a high-molecular-weight polymer by free-radical initiation, an ionic reaction must have been initiated by the high-energy radiation. Since that time many different systems of radiation-induced ionic polymerization have been found and studied.

Absolute criterions for ionic polymerizations are not too plentiful, but the dependence of the rate on the radiation intensity and on the temperature gives good indications that such polymerizations occur. Further, the effect of solvents and other additives and copolymerization reactivity ratios gives clear-cut evidence for the mechanism.

One of the intriguing discoveries about radiation-initiated ionic polymerization is the effect of water. This effect was shown in 1960 by Bates and Williams[1] but was largely overlooked. At about this same time, Bywater, in some unpublished work with α-methyl styrene, found greatly accelerated rates with radiation initiation when extremely dry monomer was used, and Stannett et al. immediately tried extreme drying conditions with isobutylene. Very fast rates were found[2] with G (monomer consumption) values up to several thousand (previously published G values varied from 300 by Collinson, Dainton, and Gillis[3] to 850 by Charlesby, Pinner, and Worrall[4]). Since reproducibility was not obtained, eventually the problem was abandoned. It was clear, however, that extremely high molecular weights resulted and that the water acted as a terminating agent. The effect of extreme dryness on the rates of radiation-induced polymerization of styrene and α-methyl styrene has been studied in a highly effective manner by Metz et al.[5-7] at Brookhaven, and Ueno, Hayashi, and Okamura[8] and their colleagues[9] at Kyoto. The problem, including isobutylene polymerization, has also been studied in detail by Williams at the University of Tennessee and during his sabbatical leave at Kyoto. A number of facets to radiation-induced ionic polymerizations which make such processes of considerable interest to

*Summary of a report by V. Stannett, E. P. Stahel, J. Kohler, E. Oda, S. Omi, A. Goineau, H. P. Shah, and F. T. Osborne, USAEC Report ORO-3667-1, North Carolina State University, June 1, 1971.

industry include (1) the possibility of obtaining extremely high G values for monomers in conventional polymerized free-radical polymerization (e.g., 600,000 for styrene at room temperature, Hayashi et al.[9]), which, coupled with purity and other aspects of radiation, could become of interest even with established systems; (2) polymerization with high G values of merely "ionic" monomers such as α-methyl styrene and isobutylene, present processes using chemical initiation often being expensive, elaborate, and not too reliable; (3) ionic polymerizations and copolymerizations in the absence of a gegenion, which may lead to unusual effects such as changes in molecular weight and molecular-weight distribution, stereochemistry, and differences in comonomer reactivity ratios and sequence distributions.

It was proposed, therefore, to conduct laboratory-scale research into new systems under superdry conditions and to construct a small pilot plant to study selected systems on a larger scale. The results of both basic laboratory and engineering studies are described in brief.

EXPERIMENTAL

Both basic laboratory studies and engineering pilot-plant research studies have been conducted on the radiation-induced polymerization of vinyl monomers under superdry conditions.

Laboratory Studies

A major part of the laboratory research was a study of various techniques for drying vinyl monomers to give optimum rates, with α-methyl styrene being used as the model monomer. A technique was developed in which baked silica gel was used for the final drying step after treatment with a series of sodium mirrors. This technique was then used with a "Gammacell 200" ^{60}Co irradiator for a number of monomer systems.

Neither trioxane—molten or in solution—nor vinyl chloride could be induced to polymerize ionically even after the most stringent drying. Dried β-pinene was found to polymerize readily but did not give yields or molecular weights greater than those already reported in the literature.

A series of α-olefins analogous to isobutylene were studied in great detail under rigorous drying conditions, but only isobutylene polymerized ionically. The other α-olefins all gave low-molecular-weight oils, probably by a free-radical mechanism. Isobutylene is believed to be unique in that it yields a stable carbonium ion and polymerizes with a very high propagation rate and with a minimum of steric hindrance. No other α-olefin appears to meet these requirements.

Finally, a study of the vinyl ethers was made, and isopropyl vinyl ether was found to polymerize at the highest yields yet reported. It was polymerized under optimum conditions for the first time, the polymerization rate having a square root dependence on the dose rate. Preliminary studies with other vinyl ethers showed a wide variety in rates with

$$\text{methyl} < \text{ethyl} < \textit{tert}\text{-butyl} < \text{isobutyl} < \text{isopropyl}$$

This order parallels closely that found with chemical carbonium-ion polymerizations. The vinyl ethers are believed to represent a most interesting series for studying the effect of structure on radiation-induced ionic polymerization.

Pilot-Plant Studies

In the second phase of the work, a recirculating-flow reactor system was used to study the radiation-induced polymerization of styrene and α-methyl styrene in bulk and in solution. The reactant monomer or monomer in solution is recirculated through an all-stainless-steel 6-liter system from a stirred vessel through relatively long transfer lines to a tubular-flow reactor positioned within a high-intensity ^{60}Co radiation field. The pilot plant is equipped with an automatic temperature-measurement and -control system for operating above and below room temperature. Constructed of standard stainless-steel tubings and fittings, the system is modular in design to facilitate changes in configuration. A polymerization-rate expression formulated from the literature was incorporated into a mathematical model for the conversion history in the system, using engineering models for each of the flow elements. The performance of the system is simulated on an analog computer. Limiting cases are solved analytically and more general cases of nonideal flow, numerically, on a large digital machine.

The flow reactor is part of a small pilot plant where both final purification and polymerization are accomplished. The ultradry moisture level and degree of conversion are continuously monitored automatically by in situ conductivity measurements and changes in refractive index.

Final purification and drying to a range below 1 part per 10^6 are accomplished on a low-temperature Molecular Sieves bed.

Studies of the dynamics of the drying process indicate that the rate-controlling step is intraparticle mass transfer. Interpretation of the rates of polymerization of styrene as a sum of a free-radical contribution and an ionic contribution is indicated, while polymerization of α-methyl styrene proceeds only by a cationic route. Nonideal viscous flow through the engineering system was studied by stimulus response techniques, and the real conversion-time behavior of the engineering flow system was characterized under various conditions. (REG)

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Puerto Rico ^{137}Cs Irradiator

By Howard T. Odum and George Drewry*

A 10-kCi ^{137}Cs irradiator, constructed by the American Nuclear Corp. of Oak Ridge, Tenn., and using ^{137}Cs from Oak Ridge National Laboratory, was transported to the El Verde rain forest by helicopter, where it was installed (Fig. 1) for studying ecological effects of radiation. The 2-ton shielded container in which the source was shipped was used as the base of the installation. It was attached to a concrete pad by wires and releasable turnbuckles.

The ^{137}CsCl source, with a density of 3.0 g/cm^3 and containing <1% ^{134}Cs, was enclosed in three stainless-steel capsules. The encapsulated source was attached to the 45-lb container plug, and the plug-source assembly was raised into the irradiation position, 6 ft above the concrete pad, by an electric motor. The main vertical tube of the installation served as a

Fig. 1 Final steps of source installation, December 1966.

*Puerto Rico Nuclear Center, Río Piedras. This article is a summary of work reported in USAEC Report TID-24270, pp. C-23-36, 1970.

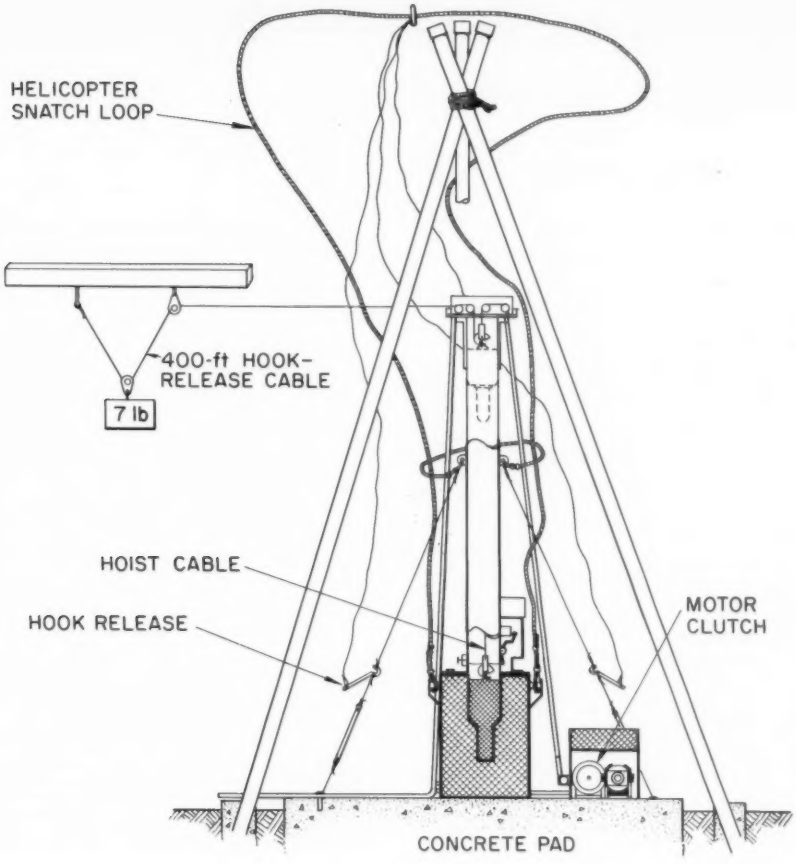

Fig. 2 Installed 10-kCi ^{137}Cs source in the El Verde rain forest.

fourth encapsulation for the elevated source. The calculated dose rate under irradiation conditions was 336 R/hr at 3 m.

A $\frac{1}{16}$-in.-diameter stainless-steel wire attached to the source hook passed through conduit, to a place 400 ft away behind a ridge, and ended in a counterweight arrangement (Fig. 2). This arrangement kept the cable taut during raising of the source. Normally, the source was raised and lowered gradually, but a scram switch allowed a quick drop in case of emergency. Silicone oil in the source well in the shielded container cushioned any sudden drops and kept out water. A tripod made of 2-in.-diameter steel pipe protected the hoisting mechanism from falling limbs, and a helicopter snatch loop was provided in case it should be necessary to remove the source quickly. (MG)

Gamma-Ray Source-Handling System*

By James Strohlein

Assisted by Louis Palombo and James McNally

A remote-handling system for a ^{60}Co gamma source, now in use by the Armed Forces Radiobiology Research Institute in Bethesda, was designed at the U. S. National Bureau of Standards, where the components were also fabricated. The system, which raises ^{60}Co slugs from a 16-ft-deep storage pool, provides for automatic return of the source to the bottom of the pool in case of mechanism failure.

The desired dose rate from the source is obtained by selecting the proper number of slugs, 30 of which can be stored in the 2.5-ft-wide trench pool. The 10-in.-long, 0.5-in.-diameter slugs are individually encapsulated in stainless steel with an outer aluminum jacket.

Two elevators for raising the slugs are mounted on two independent four-wheeled carriages, which run on tracks straddling the trench (Fig. 1). Each elevator carries a column that extends to the bottom of the trench and provides a rail to guide the source array as it moves from the pool bottom to any height up to 12.5 ft above floor level. A 21-ft-long, manually operated tool is used to load the slugs into cages, which are moved horizontally to the desired location. The objects to be irradiated are placed in the desired exposure position, and the operator leaves the room. After the shielding door has been closed, the operator raises the sources to the desired height and observes the experiment by closed-circuit television. The usual interlocks and other safety features prevent entrance to the room when the source is in the exposure position. (MG)

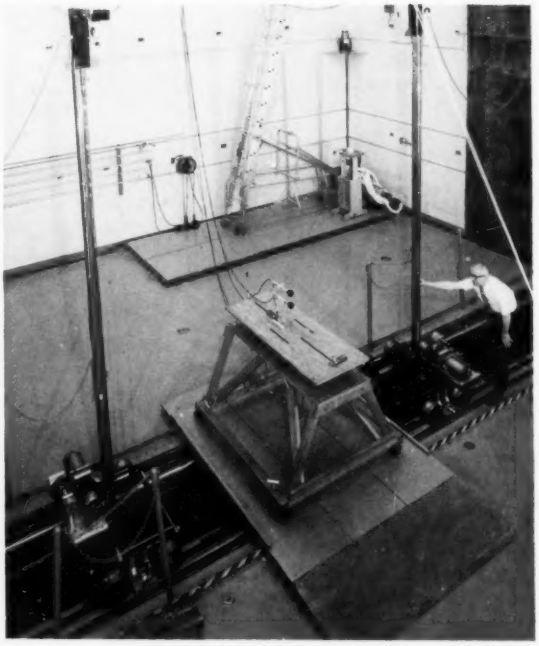

Fig. 1 NBS remote-handling system for ^{60}Co source.

*Summary of article in National Bureau of Standards *Technical News Bulletin*, **55**: 188-189 (1971).

Section V

Isotopes and Radiation Technology

Applications in Life Sciences

Vibrations in Ear Measured in Vivo by the Mössbauer Technique*

Supplementary Keywords: *medicine; physics; source, gamma; Mössbauer effect; technique; enriched stable isotope use; unusual material;* ^{57}Fe; ^{57}Co

Abstract: *At three laboratories in widely separated places—Israel,[1] Australia,[2] and the United States[3]—the Mössbauer technique has been used for in vivo measurement of the very small movements (a micron or less) of parts of the ear. In each study, the gamma-radiation source was* ^{57}Co*, and the absorber was* ^{57}Fe.

SIGNIFICANCE

The understanding of auditory physiology requires knowledge of the mechanical behavior of the cochlear partition of the ear, because its movement excites the cochlear hair cells, which are the primary sensors of acoustic energy. The transparency of the partition, its immersion in fluid, and the small distance it moves—even under great sound pressure—make the measurement of its motion very difficult, especially in living animals. The Mössbauer technique was found to be eminently suitable for this measurement.

PRINCIPLE

The Doppler effect causes the energy of a gamma ray to change when the gamma-ray source moves. A source velocity <1 mm/sec can result in an "off-resonance" shift in the gamma-ray energy if the nuclear state of the gamma energy is properly chosen. The velocity is measured by determining the degree of absorption of the gamma rays by a corresponding absorber. In this particular application of the Mössbauer effect, the gamma-ray source ^{57}Co is attached rigidly by surgery to the ear part whose motion is to be measured. The form, phase, and amplitude of the motion are measured with the fixed gamma-ray absorber ^{57}Fe, whose nucleus is identical with that of ^{57}Co; measurements in three directions determine the motion completely. The time variation of the counting rate of the gamma rays transmitted through the absorber is displayed by means of a gamma counter.

EXPERIMENTAL PROGRAMS

The three experimental programs are summarized in Table 1.

The data taken in these programs proved the usefulness of the Mössbauer technique and the satisfactory accuracy and reproducibility of results, obtained with it, over the ranges of frequency and of amplitude of biological interest. In one case, deviate data were the means to detecting a pathological distortion of the eardrum.[1] Evidence was provided that the malleus–incus system probably has no significant friction, bone flexibility, or play in the joints.[1] A value, 600 Å, obtained[2] for the maximum basilar-membrane amplitude at 90-db sound pressure and 18-kHz frequency is consistent with values obtained by another method. New information was gained about the characteristics of the ratio of the displacement of the basilar membrane to that of the malleus.[3] This ratio is useful in interpreting the mechanism of auditory response.

*For the work described in Ref. 3, Dr. W. S. Rhode received the 1970 Talbot Award of the IEEE Group on Engineering, Medicine, and Biology.

Table 1 Summary of Experimental Programs To Measure Ear Vibrations In Vivo by the Mössbauer Technique

Investigator(s)	Animals	Ear areas	14-keV gamma radiation		Sound		Measurement		
			Source	Absorber	Pressure, db	Frequency, kHz	Type	Range	Estimated Error
Gilad, Shtrikman, Hillman, Rubinstein, and Eviatar[1]	Guinea pigs	Umbo Incudostapedial joint	^{57}Co diffused in Cu (~0.3 mm dia.; 0.01 mg); 0.1 mCi	Stainless-steel foil enriched in ^{57}Fe; 1 in. dia.	~60–90	0.25–9	Amplitude phase	~6–1200 Å	±2 Å
Johnstone and Boyle[2]	Guinea pigs	Basilar membrane of first turn of cochlea Incostapedial joint	^{57}Co in stainless-steel foil (5 μ thick; 0.008 or 0.014 mm^2; 0.3 μg)	Stainless-steel foil enriched in ^{57}Fe	60–95	0.35–30	Amplitude phase	0–600 Å	
Rhode[3]	Squirrel monkeys	Basilar membrane of cochlea Bony limbus of cochlea Incus	^{57}Co on stainless-steel foil (5 μ thick; 1 mm dia.; 13 mCi) 40 by 50 μ, 60 by 60 μ, 80 by 80 μ, or 200 by 200 μ sections were used	Stainless-steel foil (rolled; 5 μ thick; 1 mg/cm^2) enriched in ^{57}Fe	70–120	0.1–25.5	Amplitude phase	~0–1000 Å	

ADVANTAGES

The Mössbauer technique has several advantages over techniques used previously. It has greater sensitivity, making possible measurements of amplitudes with an error of only ±2 Å and of velocities as small as 0.2 mm/sec. It permits measurements through small amounts of tissue. No doubt, its greatest advantage is suitability for use in vivo and the associated lack of effect of the animals' movements on the ear-vibration measurements. (HPR)

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Nuclear Engineering Applications in Medical Science

By William A. Jester,* George L. Jackson,† Bong K. Lee,*
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Supplementary Keywords: *medicine; diagnosis; therapy.*

Abstract: *The current status of nuclear medicine is reviewed, the roles that the nuclear engineer and nuclear engineering departments can play in nuclear medicine are suggested, and the specific efforts of The Pennsylvania State University's Nuclear Engineering Department in this field are described. Radioisotopes of medical interest are listed in an appendix.*

The purpose of this article is to show how nuclear engineers, working as a team with members of the nuclear medical profession, can make significant contributions toward improving the diagnostic, therapeutic, and investigative uses of radionuclides in medicine; provide improved irradiation and isotope production services; and possibly assist in nuclear medical education. Detailed discussions of nuclear medicine may be found in Refs. 1 to 4.

REVIEW OF NUCLEAR MEDICINE

The size and growth of the medical market are indicated by the fact that in 1960 the U. S. spent $\$25.9 \times 10^9$—5.1% of the gross national product—on medical care, a figure that increased to $\$67.2 \times 10^9$ (6.9%) in 1970 and is predicted to go to $\$100 \times 10^9$ by 1976 and possibly to $\$200 \times 10^9$ by the early 1980s (Refs. 5 and 6).

In 1969, about 2 thousand U. S. hospitals were using nuclear medical techniques,[7] each employing appropriate instrumentation worth, on an average, between $25 and $30 thousand. Approximately a hundred new nuclear medical facilities were being added annually, with a total annual purchase of $\$8 \times 10^6$ for appropriate instrumentation, $\$8$ to $\$10 \times 10^6$ for radiopharmaceuticals, and $\$6$ to $\$8 \times 10^6$ for radiochemicals. This does not include machine X-ray sources for radiotherapy. By 1980, nuclear instrumentation[8] alone may be a $30 to $\$50 \times 10^6$ per year market, and the total nuclear medical market, $\$100 \times 10^6$. Between 3 and 4 million patients each year benefit from medical procedures using radioisotopes, about 98% of this effort being diagnostic rather than therapeutic.[9]

Diagnostic Procedures

Imaging. The visualization of isotope distribution within the living body by scanning instrumentation has become a major medical diagnostic tool. During the last decade, the older rectilinear scanners began to be replaced by the nonmoving camera, which decreases picture exposure time and can be used to make motion pictures of the changing isotope distribution to follow

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physiological processes.[10] Combined with short-lived radioisotopes, they offer increased sensitivity and possibly increased resolution in scanning, yet reduce the patient dose.[11] The positron camera using back-to-back detectors to detect the coincidence radiation of the 0.511-MeV positrons does not require collimation, is independent of radiation attenuation, and offers the possibility of subsegment focus and of using time-of-flight measurement to give information about source depth (three-dimensional visualization).[8,9] There are some very attractive short-lived radioisotopes that decay only by positron emission—^{11}C, ^{13}N, ^{15}O, ^{18}F, and ^{30}P.

Most of the other types of cameras available use NaI(Tl) detectors—e.g., the autofluoroscope, which uses a crystal mosaic as the radiation detector; image-intensifier cameras, which use intensifiers to enhance the scintillation brilliance resulting from the absorption of photons by the detector; and image-size reduction cameras.[10]

Some work has been done on solid-state camera detectors using p–i–n junction detectors based on mosaic elements produced from Ge(Li) and Si(Li) detectors. Currently, the major problems are getting detectors large enough to have a reasonable efficiency and maintaining them at −40°C to ensure their stability.[10,12]

Another concept being investigated is the use of dynamic radiographic techniques based on the interaction of a penetrating photon field with a mechanical vibration field. Cross-correlation of the scattered radiation in two directions should make it possible not only to pick out three-dimensional images but also to derive contrast by individual mechanical response of the object's internal components—e.g., pictures of the blood system as opposed to bone or muscle.[13] Still another, called "tomography," is a method of three-dimensional scanning using specially designed collimators.

The radioisotopes more commonly used for diagnostic scanning are listed in Ref. 14. Obviously, a usable isotope must be available at reasonable cost. Further, its decay energy[10,15] must be between 80 and 400 keV—lower energies will not penetrate the intervening tissue, and, for energies higher than 400 keV, the collimators used to screen out back-scattered radiation and thus to create a sharper image are too large and inefficient. An exception to this high-energy limitation is the 0.511-MeV photon from the annihilation of positrons. Instead of a collimator, a positron camera uses a pair of detectors, one on each side of the patient, and records only the coincidence radiation of the positron pair.[9,10] Little or no particle emission is allowable—especially beta and alpha particles, which cannot be readily detected outside the body but contribute greatly to the radiation dose. Preferred isotopes are those which decay by electron capture or isomeric transition and emit only a few internal conversion electrons of very low kinetic energy. Other radionuclides formed in the production of the isotope of interest may have to be removed, or the isotope may have to be produced from an enriched target to minimize interference. For example, neutron irradiation of naturally occurring zinc will produce not only the ^{69m}Zn useful for prostate gland scanning but will also produce ^{65}Zn and ^{71m}Zn; with a target enriched to 99.3% in ^{68}Zn, the production of ^{69m}Zn is enhanced.[16] Further, a radioisotope cannot be used if its decay products interfere with the scanning—^{52}Fe, the iron isotope with the best scanning characteristics, is limited because it decays to ^{52m}Mn, which in turn decays to ^{52}Mn, both of which emit high-energy photons (1.43 MeV).[17] Finally, the radioisotope used should be of high specific activity to avoid toxic or other undesirable side effects. Such preparations may be made by chemical separation from the target or by use of isotopically enriched targets.[12,18,19]

A nontoxic form of the isotope—e.g., compound, complex, or colloid—must be available which will be taken up by the organ of interest, with little or no deposition in surrounding organs. For example, radioisotope-tagged macroaggregated albumin, which lodges temporarily in the arteriolar capillary bed, can be used for perfusion lung scanning.[20] Since strontium and fluoride ions readily deposit at bone sites where there is a rapid calcium turnover, ^{85m}Sr, ^{85}Sr, and ^{18}F can be used in bone scanning.[15] All iodine compounds are readily taken up by the thyroid. The radioisotopes used should have physical half-lives of from minutes to a few hours or be in a chemical form such that they reside in the body for only a few hours so that a larger initial amount of activity can be administered with less total absorbed dose to the patient and with a greater fraction of total radiation contributing to the scan. Repeat studies are possible in a relatively short time. A number of important elements exist in usable forms only as short-lived radioisotopes—e.g., ^{11}C, ^{13}N, ^{15}O, ^{18}F, and ^{30}P—and many short-lived radionuclides, including these five, decay by positron emission, allowing the use of the positron camera. Examples of radioisotope-labeled compounds that have a short biological half-life are orthoiodohippuric acid–^{131}I, Rose Bengal–^{131}I, and iothalamate–^{125}I.

A few examples of diagnostic imaging should be mentioned. Thyroid studies are usually conducted with rectilinear scanners to provide a life-sized display using ^{131}I (as iodine-labeled Hippuran, Rose Bengal, or albumin) or ^{99m}Tc (as pertechnetate),[21] but ^{123}I is being used increasingly as a replacement for ^{131}I. Liver and spleen imaging are done almost exclusively with nonmoving scintillation cameras using ^{99m}Tc sulfur colloid, although ^{51}Cr as the chromate is sometimes used for spleen imaging.[22] Brain studies are usually made using rectilinear scanners because of the high degree of resolution required, with ^{99m}Tc as the pertechnetate being a very popular scanning agent.[21] Pancreas imaging is accomplished primarily with ^{75}Se as selenomethionine, using a nonmoving scintillation camera.[23]

Lung imaging can be accomplished using either a rectilinear camera or a rectilinear scanner, the latter being more common. Macroaggregated albumin labeled with ^{131}I, ^{99m}Tc, or ^{113m}In is used for perfusion lung scanning and ^{198}Au colloid, ^{131}I–albumin, or ^{99m}Tc sulfur colloid for inhalation lung scanning.[10] Renal scanning has been done using ^{131}I–Hippuran and ^{203}Hg Chlormerodrin, but ^{99m}Tc-labeled compounds have been introduced recently.[8]

Bone imaging has been accomplished using ^{87m}Sr and ^{85}Sr, but ^{18}F has recently become the isotope of preference wherever it is available.[24,25]

Tracer Applications

Tracers, which must be easily and accurately detected, are used to assist in the definition of a physiological function, a compartment size, the rate of transfer from a compartment, etc. A classic example is the labeling of iodide pools with a radioactive tracer for analysis of the iodide pump mechanism, thyroxine biosynthesis, and release. A more complicated example is the labeling of an amino acid or other metabolic substrate for the purpose of determining biochemical pathways or enzymatic defects. Tracers are also used in studying circulatory phenomenons and regional blood flow.

Tracers may be either radioactive or activable, but radioactive tracers are used almost exclusively at present (Appendix 1). However, activable tracers can add a valuable dimension to tracer studies when activation capabilities are available. For example, for determination of their survival rate,[26] red blood cells can be labeled with radioactive ^{51}Cr, but the 0.32-MeV gamma ray emitted in its decay prohibits its use for children or pregnant females. However, isotopically enriched ^{50}Cr, being nonradioactive, can be used as a labeling agent and subsequently activated to ^{51}Cr in vitro,[27] eliminating all patient exposure.

Radioactive tracers may actually give less radiation exposure than did older diagnostic techniques. For example, placental localization with ^{131}I- or ^{99m}Tc-labeled albumin exposes the mother and fetus much less than the gross and often inaccurate X-ray method and with less hazard than physical examination. The newer techniques permit the accurate determination of a "central placenta previa" which, if present and not diagnosed, could result in the death of the mother and infant if normal delivery should be permitted.[28]

The radioisotopes commonly used for tracer applications are listed in Ref. 14.

In Vitro Laboratory Procedures. In order to decrease the dose of radioactivity delivered to patients, a systematic examination of means of collecting important clinical data without internal administration of radiopharmaceuticals is in progress. Progress in bench procedures includes development of radioimmunoassay techniques,[8] which primarily involve measurement of the hormones of blood, thyroxine, Parathormone, and insulin using ^{125}I and ^{131}I. This technique is elegant in its sensitivity and precision and permits accurate assay of the secretory function of most of the endocrine glands. The triiodothyronine binding, or T-3, test uses ^{131}I or, in some cases, ^{125}I.

Activation Analysis. Activation analysis, especially neutron activation analysis, is one of the most sensitive analytical methods for many elements. As mentioned above, it is sometimes used for determining isotopically enriched tracers that have been administered in stable form. However, for determination of trace elements already present, it is the preferred method or even the only method that can be used because of the possibility of contamination during the sample preparation required by other methods. Analysis may be carried out instrumentally.[29] Chemical separation, if required, can often be accomplished after sample irradiation where contamination is no longer a serious problem. Very little practical medical use is being made of activation analysis, partly because of the lack of availability of irradiation facilities and partly because of the lack of medical personnel trained to use existing facilities. But the potential of this tool is so great that it appears destined to play an important diagnostic role in medicine.

Activation analysis is, however, being used in research, e.g., in studying the distribution and function of trace elements in the human body. Of some 63 trace

elements in the human body, 7 have been identified as essential elements, and there is evidence that 7 more may be essential.[30] Activation-analysis techniques can show the limits of trace-element concentration in normal body components and the variation from these limits in abnormal samples to establish biological need or effect.[12,31-36]

An interesting possibility is the use of scalp hair and fingernail clippings for diagnostic purposes, possibly in mass screening. These two types of tissue have some ideal characteristics for screening studies in that they are painlessly removed, normally discarded, and easily collected.[37-40] Neutron activation analysis of fingernails for Na, K, Cl, and/or Cu has been proposed for mass screening of newborn infants for the early diagnosis of cystic fibrosis.[41-44] The mercury content in the hair has been shown to be related to the dietary mercury intake and thus can be used for diagnosis of mercury poisoning.[45]

Some major problems involved in this type of analysis are the contamination of such samples, especially hair, by environmental contaminants. Certain elements may accumulate in hair while the amount of the same element in other tissue is minimally or transiently elevated. Once deposited, these elements remain in an elevated concentration for many months after exposure.[38,46]

In some cases, the sample must be separated before irradiation—e.g., the four protein-bound iodine compounds in blood (MIT, DIT, T-3, and T-4) must be chromatographically separated prior to irradiation, since irradiation destroys the integrity of the molecules to such an extent that postirradiation chemical separation would show only the total iodine content of the serum.[47,48] Vitamin B_{12} can be determined in blood by postirradiation separation, since the activation products sought—^{60}Co and ^{60m}Co—are formed by neutron activation of cobalt, and the only significant amount of cobalt in the blood is in the form of vitamin B_{12} (Ref. 49).

Another type of neutron activation procedure that has not yet found much application for the analysis of biological samples is neutron-capture gamma analysis, where the prompt capture gammas are measured as the sample is being irradiated. This method offers the potential of analyzing for some elements that are difficult to determine in more conventional ways. One author reports[50] being able to analyze biological samples for H, B, N, Na, S, Cl, K, Ca, Ni, Cu, Ge, and Hg.

Not only in vitro but also in vivo activation-analysis procedures are being investigated for the determination of the content of certain elements in the whole body or in a localized region, such as the thyroid gland. Irradiations are usually done with high-energy beams of neutrons from accelerators or pulsing reactors in order to even out the flux distribution throughout the body. Radiation doses to the patient are no greater than those incurred during an X-ray examination, and, with a conventional whole body counter, the total body contents of Ca, Na, Cl, N, and P and the iodine content of the thyroid can be determined.[51-54]

Another interesting in vivo application is the recent development of a ^{252}Cf focused neutron gun for the activation of skin tissue to detect above-normal amounts of sodium for the diagnosis of cystic fibrosis.[55]

Therapeutic Procedures

The isotopes commonly used for therapeutic applications are listed in Ref. 14. The techniques used include radiopharmaceutical use, brachytherapy, teletherapy, and extracorporeal irradiation.

Radiopharmaceutical therapy involves the administration of a radioisotope to deliver a therapeutic dose to a specific body organ or to a diseased portion thereof. Unlike radioisotopes chosen for scanning applications, it is desirable that the chosen radioisotope emit high-energy charged particles to obtain the desired effect. For example, the treatment of hyperthyroidism with ^{131}I is now considered by many to be the treatment of choice, and surgical treatment has decreased phenomenally over the past two decades. Thyroid cancer is thought to be best treated by a combined surgical–radiopharmaceutical approach. Certain other cancers are treated[56,57] with ^{32}P-, ^{198}Au-, and ^{90}Y-labeled radiopharmaceuticals.

Brachytherapy—the interstitial placing of radioisotopes in diseased areas of the body—includes the implantation[58] of wires or needles containing beta- or high-energy gamma-emitting isotopes such as ^{60}Co, ^{198}Au, and ^{192}Ir. An interesting application of ^{252}Cf is in tumor implants for anoxic (low-oxygen) tumors, since fast neutrons are the only effective and practical source of radiation currently available. The actual particles that irradiate the tumor are recoil protons caused by the collision of the neutrons with hydrogen atoms in and near the tumor. Thus ^{252}Cf implants have a great potential in the treatment of such tumors.[59]

The main radioisotopes for gamma-ray teletherapy—where the source is at a distance from the patient—are ^{60}Co and ^{137}Cs, which are used not only

to treat cancer but also to suppress the immune reaction involved in organ transplants.[8] High-energy neutrons from cyclotrons are often used for the treatment of anoxic tumors,[60] but thermal neutrons can be used if the tumors are saturated with ^{10}B or ^{6}Li, since alpha particles are produced in the $^{10}B(n,\alpha)^{7}Li$ and $^{6}Li(n,\alpha)^{3}H$ reactions.[18] Beams of heavy ions and negative pi-mesons may in the future be used to treat anoxic tumors.[8]

Extracorporeal irradiation has been used in leukemia therapy, blood and lymph being irradiated outside the body using beta and/or gamma sources. The blood flows from an artery into a plastic tube that passes through a high-intensity radiation field and then back into a vein. Only the blood cells are affected; the rest of the body receives a minimal dose. The results obtained are similar to those obtained from the drugs used in leukemia treatment but without their toxic side effects. This technique is also being used in suppression of immune antibody response to organ transplants and grafts.[8,61]

Isotopic Power Sources

The major radioisotope being considered as an implantable energy source for biomedical applications is 87.4-year ^{238}Pu. It emits only 5.5-MeV alpha particles (the source of the heat) and easily shielded low-energy photons. For biomedical purposes it must have less than 0.1 ppM of ^{236}Pu, which emits unwanted gamma rays. If the ^{238}Pu isotope is in the oxide form, the oxygen must be depleted in ^{17}O and ^{18}O to avoid the production of (α,n) neutrons.[62]

The heat of decay can be converted to electrical energy to power devices such as heart pacemakers or to provide mechanical power for artificial organs.[8] It is predicted[63] that 60 g of ^{238}Pu could run a heart pump for 90 years, supplying variable power levels of from 1.7 to 7 W.

Sources of Radioisotopes for Nuclear Medical Applications

Various methods are used to make available the widest possible range of radioisotopes for medical applications. An example of a naturally occurring radioisotope used in medical studies is ^{40}K, found in all potassium, with which the lean body mass under normal development and body-wasting conditions can be determined. Because only small amounts of radiation are involved, whole body counters are used.[8] However, most radioisotopes used in medicine are produced artificially.

Thermal-Neutron-Produced Radioisotopes. Usually produced in nuclear reactors, these radioisotopes are generally beta producers and thus are often not suitable for medical applications because of the body doses involved. However, the activation products of lower-atomic-weight isotopes of an element frequently decay by electron capture, isomeric transition, or low-energy positron decay, and, if the irradiated sample is enriched with this isotope, a suitable product is formed—e.g., ^{51}Cr(EC), ^{55}Fe(EC), ^{69m}Zn(IT), ^{85m}Sr(IT), and ^{87m}Sr(EC). If the radioisotope decays to a radioisotope of a different element that has suitable characteristics, it can usually be chemically separated[8,16] from interfering activities in the carrier-free form—e.g., ^{47}Sc from ^{47}Ca and ^{131}I from ^{131}Te.

There is also the possibility of using the neutron to produce secondary charged particles for radioisotope production. A classic example is the production of the important bone-scanning isotope ^{18}F by neutron irradiation of ^{6}Li-enriched lithium carbonate by the reactions $^{6}Li(n,t)^{4}He$ and $^{16}O(t,n)^{18}F$. Here the triton produced from the neutron capture in ^{6}Li irradiates[64] the oxygen to produce a high yield of the 110-min positron-emitting ^{18}F.

Fast-Neutron-Produced Radioisotopes. Fast-neutron reactions are usually of the (n,p) and (n,α) types, and the product isotope, being chemically different from the target material, is easily separated chemically. Examples[16] are $^{33}S(n,p)^{33}P$, $^{43}Ca(n,p)^{43}K$, $^{67}Zn(n,p)^{67}Cu$, and $^{69}Ga(n,p)^{69m}Zn$. The yield of these reactions is usually much lower than the (n,γ) thermal-neutron reactions because of smaller cross sections.

Accelerator-Produced Radioisotopes. Accelerators, especially cyclotrons, are used to produce a wide variety of radioisotopes, which are usually neutron deficient and thus often decay by electron capture or positron emission. The types of bombarding particles are usually protons, deuterons, tritons, ^{3}He, and alpha particles. Some typical examples[8,16] are $^{67}Zn(p,2n)^{66}Ga$, $^{5,6}Fe(d,n)^{57}Co$, $^{84}Sr(p,2p)^{83}Rb$, and $^{50}Cr(\alpha,2n)^{52}Fe$. Of special interest are the low-atomic-weight short-lived positron-emitting radioisotopes that can be produced by small cyclotrons located at medical establishments. Some of the more important of these reactions[8,11,20] and their half-lives are $^{11}B(p,n)^{11}C$ (20 min); $^{10}B(\alpha,n)^{13}N$ (10 min); $^{14}N(d,n)^{15}O$ (2 min); $^{18}O(^{3}He,p)^{18}F$ (110 min); $^{27}Al(\alpha,n)^{30}P$ (25 min); and $^{35}Cl(\alpha,n)^{38}K$ (7.7 min).

Fission Products. Fission products, which have an excess of neutrons and therefore usually decay by beta-particle emission, have found some use in medical applications. For example, the fission gases ^{85}Kr and ^{133}Xe are used to measure precise volume of blood flow in specific areas of the brain.[8] There is a short physical half-life of these gases in the body since they are swept out of the blood by the lungs, thus lowering the dose resulting from the beta particles they emit. Fission product ^{90}Sr and its daughter ^{90}Y are used as extracorporeal irradiators of blood and lymph fluids,[61] and ^{137}Cs is a major isotope for teletherapy.[8,17]

Fission products of high yield and long half-life can also be irradiated to produce radioisotopes of more desirable characteristics than the product itself. For example, the fission product ^{129}I with a 1.6×10^7-year half-life (cross section of 28 barns) can be irradiated with thermal neutrons to produce 12.3-hr ^{130}I, and the unique radiation produced gives it the potential of being an excellent activable iodine tracer.

Transuranium Isotopes. The potential availability of relatively large amounts of transuranium isotopes from power reactors and high-flux reactors is leading to the investigation of these radioisotopes for medical application. These isotopes are produced mainly by multiple neutron capture in ^{238}U. Some current applications are ^{238}Pu as an isotopic power source for heart pacemakers and possibly artificial organs,[62] ^{242}Am as an X-ray-fluorescence source for in vivo analysis,[65] and the intense neutron source ^{252}Cf for tumor implants, teletherapy, and medical neutron radiography.[8,12,66]

Radioisotope Generators. An important source of short-lived radioisotopes which has recently become available for nuclear medical applications is the radioisotope generator, sometimes called a "cow," involving a radioisotope that decays to a daughter of significantly shorter half-life. The parent isotope is chemically attached to a substrate, such as an ion-exchange resin, and the daughter isotope is eluted or "milked" off the substrate by a solvent that is selective for the daughter. Thus the short-lived daughter can be used at great distances from the original site of production.[21] Between elutions, the activity of the daughter product builds back up in the substrate, reaching an equilibrium value with the parent after a time equal to a few half-lives of the daughter. The parent radioisotope may come from any of the previously mentioned sources of production. Examples of currently available isotope generators are

Parent	Daughter
^{132}Te (78 hr)	^{132}I (2.3 hr)
^{99}Mo (67 hr)	^{99m}Te (6.0 hr)
^{113}Sn (115 days)	^{113m}In (1.7 hr)
^{87}Y (80 hr)	^{87m}Sr (2.8 hr)
^{68}Ge (275 days)	^{68}Ga (58 min)

Research is currently under way to study the potential application of isotope generators that produce extremely short-half-life radioisotopes to be used with high-speed scanning equipment for dynamic studies.[67] Some examples are

Parent	Daughter
^{109}Cd (453 days)	^{109}Ag (39 sec)
^{191}Os (15 days)	^{191m}Ir (4.9 sec)
^{77}Br (57 hr)	^{77m}Se (18 sec)

Problems and Challenges of the Nuclear Medical Field

There is a need to develop techniques for mass screening of large numbers of people for early diagnosis of disorders and for preventive maintenance as a means of increasing the physical well-being of the public at large.[6] Radioisotope scanning is a powerful tool in diagnostic medicine although the method currently is relatively crude. Even though most of the organs of the body can be scanned to some degree, tumors or cancers cannot be detected in their early growth (a tumor must contain about 10^9 cells to be detected). Radioactive tracers and detectors that can detect as few as 10^6 cells are needed. It would then be possible to remove the tumor before it has a chance to spread to other areas of the body and would lead to greatly increased survival rates of cancer patients.[12]

The inherent picture capabilities of current imaging devices is adequate, but their usefulness is often limited by poor counting statistics and/or by low contrast. These result from low isotope concentration within the area of interest and the use of less than the optimum amount of radioactive material in deference to patient dose considerations. The problem and challenge is in making available short-lived isotopes that will allow increased source strength and more specific vehicles for the deposit of these isotopes in the organ of interest.

Carbon-11, with its 20.5-min half-life, is not yet usable outside of research applications. It is potentially

the most important single radioisotope, since carbon is the basic building block of the human body. Unlimited chemical compounds tagged with a "scannable" carbon that is readily available would pave the way for a multitude of diagnostic studies.

Another problem in imaging devices is that current picture-processing methods fail to extract the full amount of information. Much could be accomplished with computer processing of the data.[10]

There is also the problem of increasing the genetically significant dose to the general population. Estimates based on a 1964 study indicated an annual dose of 55 mR per person from X-ray examinations alone. Current estimates put this level at about 90 mR per person. It is estimated that, even taking into account improved inspection procedures and improved care in the applications of X rays, by 1990 the annual genetically significant dose could be as high as 200 to 400 mR per person.[7]

The public can see more clearly the risk vs. benefit involved with increased radiation exposure from the use of nuclear medical techniques, since these techniques have greatly improved the quality of patient care. In some instances the use of newer nuclear diagnostic techniques actually decreases the radiation exposure by eliminating the need for making X rays, as in the case of the previously mentioned placental localization.

Currently, nuclear medicine, like so many areas of medicine, is facing the transitional growing pains as a bureaucracy concerned with medical delivery systems begins to incorporate radiopharmaceuticals within the purview of the Food and Drug Administration. The concepts of radiation and radiation technology are not easily translated, understood, or implemented by conventional pharmacists, and this creates a problem that may not be solved but should at least be understood by those becoming involved in this field.

Another problem seems to be the reluctance of pathologists to accept the newer in vitro diagnostic procedures involving the use of radioisotopes and, when it is available, the diagnostic capability of activation analysis.

As in most areas of medicine, there is a training problem—the developing need in nuclear medicine is for nuclear medical technicians.[7] There are currently about 5500 nuclear medical technicians in the United States, but by 1976 there will be a need for about 12,500, and by 1980 about 17,000. Hospitals are not equippped to handle the training of such a large expansion of technicians, and they can no longer justify tying such medical education to the patient's cost. Most of the increased need for education of nuclear medical technicians must be met by public educational institutions.

ROLE OF NUCLEAR ENGINEERING IN NUCLEAR MEDICINE

Medical Services

Because of his training, the nuclear engineer is the logical individual to supply radioisotopes for medical use. He should be in charge of the strategically located reactors and accelerators needed to supply radioisotopes with the desired radiation-emission properties, half-lives, and chemical properties, and his engineering background should lead to economic methods of production and delivery of such isotopes. His contacts with pharmacists would lead to the design and production of new and exciting combinations of short-half-life nuclides with various chemical vehicles in order to produce a degree of organ or functional specificity that is desirable but currently unavailable.

The same reactors and/or accelerators required for the production of clinical and therapeutic radioisotopes could also provide diagnostic activation-analysis services to augment existing methods of clinical analysis. Here the nuclear engineer needs the assistance of a valuable colleague—the radiochemist—since most activation analysis of biological samples requires either preirradiation separation or postirradiation radiochemistry.

Due to the wide variation in physical characteristics of patients and the ever-growing number of usable isotopes, the nuclear engineer, in conjunction with nuclear medical personnel, could play a role in determining the type and amount of radioisotope to be used in a particular case. He could also specify the amount, type, and location of the shielding to be used and conduct radiation monitoring to ensure that only the minimum required dose is administered to the patient.

Because of his background in the use of radiation detection, nuclear instrumentation, and data processing, the nuclear engineer could play an important role in ensuring that the conditions for each examination requiring the use of nuclear instrumentation and radioisotopes are such that the maximum amount of information is obtained to aid the physician in his diagnosis. The nuclear engineer could also play an important role in overseeing the maintenance of the nuclear medical instrumentation to ensure its continuously reliable operation.

Nuclear Medical Research and Development

The nuclear engineer's association with nuclear medical personnel should lead him to the identification of radionuclides with the potential for solving particular problems that his medical colleagues encounter. Having identified such radionuclides, his background should aid him in producing enough of them for a fair medical evaluation. Then, if a radioisotope proves successful, the nuclear engineer would further develop the methods of production and delivery so that a high-quality product could be made available to physicians at a reasonable price.

The use of clinical activation analysis is just in its infancy. Wide areas of research are still needed, especially in identifying essential elements and in determining the normal range of trace elements in the various organs of the human body. Of the many analytical techniques, activation analysis offers the best combination of a high degree of sensitivity and freedom from analytical contaminants. Once a suitable analytical technique has been determined which allows a diagnosis because of elemental variations from the norm, work will be required to make the technique rapid and available while minimizing the cost.

The use of nonradioactive but activable tracers should play an ever-increasing role in medicine as the public becomes more concerned about radiation dosage.

Development of New Instruments, Devices, and Techniques

Nuclear engineers, especially those having an electronics bent, can play an important role in the rapid introduction of the latest techniques, such as solid-state technology, into the nuclear medical field. Little use has been made to date of the high-resolution Ge(Li) and Si(Li) detectors. Likewise, in the area of radiation collection and data processing, little use has been made of coincidence techniques and on-line computer data processing, and virtually no use has been made of cross-correlation techniques.

The whole problem of shielding both the patient and hospital personnel from undesirable radiation fields presents the nuclear engineer with a variety of shielding problems. Methods are needed to protect the more radiation-sensitive organs during radiation therapy or diagnostic studies. The use of shielding solutions such as liquid baths or internal shielding could result from research in these areas.

ROLE OF NUCLEAR ENGINEERING DEPARTMENTS IN NUCLEAR MEDICINE

Historically, most of the interaction between the nuclear engineer and members of the medical profession has taken place at the National Laboratories—notably Brookhaven, Argonne, and Oak Ridge—through their medical divisions. Little has been accomplished at the university level to bring about the joining of nuclear engineering and the medical disciplines, although some exceptions include the cooperation of the nuclear engineering personnel from MIT with Massachusetts General Hospital; Texas A&M University with Baylor University School of Medicine; the University of Washington with the Medical Center of Seattle; and the University of Missouri with its College of Medicine. In most nuclear engineering programs, there appears to be little emphasis on the opportunities and challenges to the students outside the nuclear power field and little opportunity for research in nonpower fields. Through course work, research, seminars, and direct contact with nuclear medical personnel, the nuclear engineering student can be shown that his background in nuclear engineering can enable him to make valuable contributions to the medical field.

Nuclear engineering departments can also help nuclear medical personnel. The education of the physician will have little significant relation to nuclear engineering unless the student in his graduate education elects to enter the field of nuclear medicine. At this point his training can receive considerable supplementation from departments of nuclear engineering. Most hospitals that have the resources and expertise for clinical training do not have the necessary support for providing the basic nuclear concepts. The facilities and faculty of nuclear engineering departments could be used to meet this educational need.

Further, in the matter of medical education, there is the need to provide personnel with the technical skills for the operation of clinical services in nuclear medical laboratories. These technologists will be primarily 2-year associate-degree graduates with 6 months to a year of training in medical institutions affiliated with the university for this purpose. Nuclear engineering departments could cooperate in the awarding of such associate degrees.

The interaction of the faculty of nuclear engineering departments with nuclear medical personnel could lead into many areas of important, interesting, and challenging research mentioned above. Student involve-

ment in such research will also widen their vision as to the available opportunities.

The nuclear engineering departments could use their nuclear facilities to provide services to the medical organizations in their areas. These services could include the production of short-lived radioisotopes, such as ^{18}F, which are currently not widely available. They could also provide activation-analysis services such as the proposed method of screening newborn infants for cystic fibrosis by means of fingernail analysis. In days of tight money, such services would do much to help justify the high cost of supporting irradiation facilities such as reactors or accelerators.

Nuclear Medical Efforts of the Nuclear Engineering Department at The Pennsylvania State University

Over a period of years several of the authors of this article have been planning the formation of a nuclear biomedical team of nuclear engineering and nuclear medical specialists. Thanks to a grant provided by the Pennsylvania Science and Engineering Foundation, such a team was formed in June 1970 and has become a vital and active entity. The initial objectives of this program were to establish procedures for a nuclear biomedical team which could be applicable to doctors and medical research workers and to make available and develop new uses for clinical neutron activation analysis. More recently, additional objectives have been to provide short-lived radioisotopes for nuclear medical applications at hospitals near Penn State's Breazeale Nuclear Reactor Facility and to develop a ^{252}Cf irradiator with a potential of providing activation-analysis, isotope-production, and neutron-therapy capability for medical organizations.

Areas of effort involving the nuclear biomedical team include:

1. Decrease of sodium and chlorine interference in activation analysis of biological samples.

2. Monitoring of laboratory water quality and establishment of a capability to provide such a service to medical laboratories and users of dialysis machines throughout the state of Pennsylvania.

3. Study of sample contamination during sample collection.

4. Determination of serum calcium for diagnostic purposes.

5. Study of the relation between zinc levels and behavioral problems and mental retardation.

6. Measurement of serum T-3, T-4, and possibly mono- and di-iodotyrosine in human serum.

7. Evaluation of instrumental copper and sodium determination in fingernail samples as a potential method for the identification of cystic fibrosis.

8. Measurement of trace elements in the amniotic fluid of mothers who have given birth to children with cystic fibrosis and in those who have had normal pregnancies.

9. Measurement of cobalt in serum for determination of vitamin B_{12}.

10. Comparison of trace-element concentrations in lung tissue of individuals having respiratory disorders with those in normal lung tissue.

The major effort currently under way is the production of ^{18}F as a bone-scanning agent by irradiation of ^{6}Li-enriched lithium carbonate. Among other isotopes of interest in production are ^{85m}Kr and ^{87}Kr for blood-flow studies, and ^{198}Au and ^{194}Ir ribbons or wires for removable or permanent tumor implants.

It is hoped that in the near future a series of intense ^{252}Cf sources can be obtained from the AEC's Savannah River Laboratory to start the development of an assembly that would be the prototype of a simple irradiation device for installation at hospitals for use in activation analysis, isotope production, and neutron therapy.

Members of Penn State's Nuclear Engineering Department have teamed up with members of the University's Biomechanics Laboratory and are using their skills in nuclear instrumentation and data processing to develop improved medical instrumentation. One project, supported by the National Institutes of Health's Science Support Program, concerns the development of a gamma-ray scanning system that allows the study of human-body-segment mass parameters such as centers of gravity and moments of inertia. This system and other electronic scan systems based on the same principles are expected to aid researchers immeasurably in human performance and possibly to lead to improved design of artificial limbs. A second project is the enhancement, modulation, containment, and focusing of thermal-neutron beams using vibrating crystal diffractors, an eventual medical use of which would be in tumor radiation therapy in conjunction with and without boron saturation. And a third project is the development of dynamic radiography techniques that would utilize the interaction of a penetrating photon field with a mechanical vibration field. By

cross-correlation of the resulting scattered radiation in two directions, three-dimensional pictures of blood vessels and muscle–ligament systems, as compared to bones, might be obtainable.

Other than a brief introduction to nuclear medicine in a course called "Radionuclear Concepts" and some coverage in "Radiological Safety," there are currently no formal courses in which nuclear medicine interacts with nuclear engineering. Several graduate students are involved in the departmental research described above. Conversations are under way between members of the Nuclear Engineering Department faculty and the heads of nuclear medicine departments of some of the larger hospitals in central Pennsylvania on how efforts can best be combined to implement programs of nuclear medical education. (MG)

Appendix I Radioisotopes of Medical Interest

Radio-isotope	Production methods	Half-life	Max. energy of major decay, MeV	Major photon energies, MeV	Some medical applications	Comments	References
^{3}H	$^{6}Li(p,\alpha)^{3}H$	12.2 years	β^- (0.019)	No photon	Body water		68
^{11}C	$^{11}B(p,n)^{11}C$	20 min	β^+ (0.97)	0.511	Lung and heart function	Requires in-hospital accelerator; rapid chemistry required to form compounds	21, 12, 8
^{13}N	$^{14}N(p,pn)^{13}N$	10 min	β^+ (1.20)	0.511	Lung	In-hospital accelerator	21, 8
^{15}O	$^{14}N(d,n)^{15}O$	2 min	β^+ (1.74)	0.511	Lung	In-hospital accelerator	8, 9, 11
^{18}F	$^{6}Li(n,t)^{4}He$, $^{16}O(t,n)^{18}F$, $^{16}O(^{3}He,p)^{18}F$	110 min	β^+ (0.64)	0.511	Bone scanning	Enriched target	8, 11, 21, 24, 69
^{22}Na	$^{19}F(\alpha,n)^{22}Na$	2.62 years	β^+ (0.55)	0.511, 1.28	Body water, sodium space		70
^{24}Na	$^{23}Na(n,\gamma)^{24}Na$	15 hr	β^- (1.39)	1.37, 2.75	Sodium space		
^{28}Mg	$^{26}Mg(t,p)^{28}Mg$	21 hr	β^- (0.46)	0.031, 0.40, 0.95, 1.35	Magnesium absorption	Only practical Mg tracer but gives ^{28}Al	17
^{30}P	$^{27}Al(\alpha,n)^{30}P$	2.5 min	β^+ (3.24)	0.511		In-hospital accelerator	21
^{32}P	$^{31}P(n,\gamma)^{32}P$, $^{29}Si(^{3}He,np)^{32}P$	14 days	β^- (1.71)	No photon	Platelet survival, lung cancer therapy		71
^{33}P	$^{33}S(n,p)^{33}P$	25 days	β^- (0.248)	No photon			16
^{38}K	$^{35}Cl(\alpha,n)^{38}K$	7.7 min	β^+ (2.68)	0.511, 2.17		In-hospital accelerator	21
^{40}K	Naturally occurring				Lean body mass studies		8
^{42}K	$^{41}K(n,\gamma)^{42}K$	12.5 hr	β^- (3.52)	1.52	Electrolyte space		
^{43}K	$^{43}Ca(n,p)^{43}K$, $^{43}Ar(^{3}He,np)^{43}K$	22 hr	β^- (0.83)	0.373, 0.39, 0.59, 0.619	Tumor scanning, blood flow, red-cell labeling	Enriched target, fission-product activation	72
^{45}Ca	$^{44}Ca(n,\gamma)^{45}Ca$	165 days	β^- (0.252)	No photon	Calcium absorption		16, 73
^{47}Ca	$^{46}Ca(n,\gamma)^{47}Ca$	4.5 days	β^- (0.67)	1.31	Calcium absorption		74
^{49}Ca	$^{48}Ca(n,\gamma)^{49}Ca$	8.8 min	β^- (1.95)	3.10, 4.1	Activable tracer	Enriched target isotope	18
^{47}Sc	$^{46}Ca(n,\gamma)^{47}Ca \xrightarrow[4.5\ days]{\beta^-} {}^{47}Sc$	3.4 days	β^- (0.60)	0.160			16
^{51}Cr	$^{50}Cr(n,\gamma)^{51}Cr$, $^{51}V(p,n)^{51}Cr$	28 days	EC	0.320	Spleen scanning, red-cell survival, tumor implants, RBC	Activable tracer, enriched target isotope	8, 18 22, 26
^{52}Fe	$^{52}Cr(^{3}He,3n)^{52}Fe$	8 hr	β^+ (0.80)	0.165, 0.511	Metabolism, bone-marrow function	In-hospital accelerator, best iron tracer, ^{52}Mn daughter	11, 17

Appendix I (Continued)

Radio-isotope	Production methods	Half-life	Max. energy of major decay, MeV	Major photon energies, MeV	Some medical applications	Comments	Refer-ences
^{55}Fe	^{54}Fe$(n,\gamma)^{55}$Fe	2.6 years	EC	Mn X rays	Iron absorption and incorporation in blood	Enriched target	8, 18, 75
^{59}Fe	^{58}Fe$(n,\gamma)^{59}$Fe	45 days	β^- (0.475), EC	1.09, 1.29	Iron absorption and incorporation in blood	Enriched target	18
^{57}Co	^{56}Fe$(d,n)^{57}$Co	270 days	EC	0.122, 0.136	Vitamin B_{12}	Preferred over ^{60}Co	8, 76
^{60}Co	^{59}Co$(n,\gamma)^{60}$Co	5.2 years	β^- (0.314)	1.17, 1.33	Vitamin B_{12} teletherapy, suppressing immune reactions		8, 18
^{64}Cu	^{63}Cu$(n,\gamma)^{64}$Cu	13 hr	EC, β^- (0.57) β^+ (0.66)	0.511	Copper metabolism		18, 19
^{67}Cu	^{67}Zn$(n,p)^{67}$Cu	58 hr	β^- (0.57)	0.092, 0.184	Copper metabolism	Enriched target	16, 19
^{65}Zn	^{64}Zn$(n,\gamma)^{65}$Zn	245 days	β^+ (0.327)	0.511, 1.115	Wound healing		77
^{69m}Zn	^{69}Ga$(n,p)^{69m}$Zn, ^{68}Zn$(n,\gamma)^{69m}$Zn	13.8 hr	IT	0.439	Prostate gland scanning	^{69}Zn daughter target	16
^{66}Ga	^{67}Zn$(p,2n)^{66}$Ga	9.5 hr	β^+ (4.15), EC	0.511, 1.04, 2.75	Lymphoid tumors and bone scanning		16, 17
^{67}Ga	^{68}Zn$(p,2n)^{67}$Ga	78 hr	EC	0.093, 0.184, 0.296	Soft-tissue tumor and metastasis scanning		8, 11, 17
^{68}Ga	^{66}Zn$(\alpha,n)^{68}$Ge $\xrightarrow[2.4\ \text{hr}]{\beta^+,\ \text{EC}}$ ^{68}Ga	68 min	β^+ (1.90), EC	0.511	Lung scanning	Isotope generator	11, 21
^{74}As	^{74}Ge$(p,n)^{74}$As	18 days	β^+ (0.95), β^- (1.36), EC	0.511, 0.596, 0.635	Brain scanning, study wound healing	Enriched target	18, 77, 78
^{73}Se	^{72}Ge$(^3$He$,2n)^{73}$Se	7.1 hr	β^+ (1.30)	0.066, 0.359, 0.511	Sulfur substitute	In-hospital accelerator	
^{77m}Se	^{75}As$(\alpha,2n)^{77}$Br $\xrightarrow[57\ \text{hr}]{\text{EC}}$ ^{77m}Se	18 sec	IT	0.161	Dynamic studies	Isotope generator	67
^{82}Br	^{81}Br$(n,\gamma)^{82}$Br	35 hr	β^- (0.44)	0.554, 0.619, 0.698, 0.777, 0.828, 1.04, 1.31, 1.48	Bladder tumor therapy	Enriched target	18
^{85}Kr	Fission product	10 years	β^- (0.67)	0.514	Blood flow, body water		8, 79
^{81}Rb	^{81}Br$(^3$He$,3n)^{81}$Rb	4.7 hr	EC, β^+ (1.03)	0.253, 0.450, 0.511	Splenic volume	In-hospital accelerator	
^{83}Rb	^{84}Sr$(p,2p)^{83}$Rb	83 days	EC	0.53	Muscular dystrophy		17
^{84}Rb	^{84}Kr$(p,n)^{84}$Rb	33 days	EC, β^+ (1.66)	0.511, 0.88	Myocardium scanning	Used with positron camera	17
^{86}Rb	^{85}Rb$(n,\gamma)^{86}$Rb	18 days	β^- (1.78)	1.078	Myocardium scanning, sodium space		80
^{85m}Sr	^{84}Sr$(n,\gamma)^{85m}$Sr	70 min	IT	0.150, 0.231	Bone scanning	Enriched target	9, 18, 72
^{85}Sr	^{84}Sr$(n,\gamma)^{85}$Sr	64 days	EC	0.514	Bone scanning and lesions	Enriched target	18, 25
^{87m}Sr	^{86}Sr$(d,n)^{87}$Y $\xrightarrow[80\ \text{hr}]{\text{EC}}$ ^{87m}Sr, ^{86m}Sr$(n,\gamma)^{87m}$Sr	2.8 hr	IT	0.388	Metabolism, bone scanning		8, 21, 25

(Table continues on the next page.)

Appendix I (Continued)

Radio-isotope	Production methods	Half-life	Max. energy of major decay, MeV	Major photon energies, MeV	Some medical applications	Comments	Refer-ences
^{90}Y	^{90}Sr (fission product) $\xrightarrow[28\text{ years}]{\beta^-}$ ^{90}Y	64 hr	β^- (2.27)	No photon	Extracorporeal irradiation of blood and lymph		61
^{99m}Tc	^{98}Mo$(n,\gamma)^{99}$Mo $\xrightarrow[67\text{ hr}]{\beta}$ ^{99m}Tc	6.0 hr	IT	0.140	Perfusion, thyroid, salivary glands, brain scanning, aerosol lung scanning	Enriched target, isotope generator, most popular scanning agent	8, 9, 11, 18, 20, 21, 81
^{109m}Ag	^{108}Cd$(n,\gamma)^{109}$Cd $\xrightarrow[453\text{ days}]{\text{EC}}$ ^{109m}Ag	39 sec	IT	0.088	Dynamic studies	Isotope generator	44
^{111}In	^{112}Cd$(p,2n)^{111}$In	2.8 days	EC	0.173, 0.247	Soft-tissue tumor scanning	Enriched target	17
^{113m}In	^{112}Sn$(n,\gamma)^{113}$Sn $\xrightarrow[115\text{ days}]{\text{EC}}$ ^{113m}In	100 min	IT	0.393	Bone scanning, perfusion studies, brain scanning	Enriched target, isotope generator	9, 11, 18, 20, 21
^{123}I	^{123}Te$(p,n)^{123}$I, ^{126}Te$(^3$He$,pn)^{123}$I	13 hr	EC	0.159	Thyroid function, blood metabolic studies	In-hospital accelerator	11, 16
^{125}I	^{124}Xe$(n,\gamma)^{125}$Xe $\xrightarrow[17\text{ hr}]{\text{EC}}$ ^{125}I, ^{123}Sb$(\alpha,2n)^{125}$I	60 days	EC	0.035	T-3 test, radioimmuno-assay, miniature X-ray source, plasma volume, kidney function	Preferred over ^{131}I for scanning	8, 82, 83
^{130}I	^{129}I$(n,\gamma)^{130}$I	12 hr	β^- (1.04)	0.419, 0.538, 0.669, 0.743, 1.15	Activable tracer	^{129}I fission product	84
^{131}I	^{130}Te$(n,\gamma)^{131}$Te $\xrightarrow[25\text{ min}]{\beta^-}$ ^{131}I	8.1 days	β^- (0.61)	0.364, 0.637	Perfusion studies, thyroid, aerosol lung scanning, T-3 and T-4 tests, fat absorption, kidney function	Enriched target	8, 9, 18, 20
^{132}I	^{132}Te (fission product) $\xrightarrow[78\text{ hr}]{\beta^-}$ ^{132}I	2.3 hr	β^- (2.12)	0.52, 0.67, 0.77, 0.955, 1.40	Thyroid, placental localization, circulation	Isotope generator	21, 28
^{129}Cs	^{127}I$(\alpha,2n)^{129}$Cs	32 hr	EC	0.375, 0.416	Stomach emptying		16
^{137}Cs	Fission product	30 years	β^- (0.514)	0.662	Teletherapy, suppressing immune reactions		8, 18
^{133}Xe	Fission product	5.3 days	β^- (0.35)	0.081	Blood flow		8, 85
^{135m}Ba	^{134}Ba$(n,\gamma)^{135m}$Ba	29 hr	IT	0.268	Bone scanning	Enriched target	72
^{140}Pr	^{141}Pr$(p,2n)^{140}$Nd $\xrightarrow[3.3\text{ days}]{\text{EC}}$ ^{140}Pr	3.4 min	EC, β^+ (2.32)	0.511			7, 17
^{153}Sm	^{152}Sm$(n,\gamma)^{153}$Sm	47 hr	β^- (0.076)	0.022, Eu L X rays	Bone scanning		69
^{171}Er	^{170}Er$(n,\gamma)^{171}$Er	7.5 hr	β^- (1.06)	0.112, 0.296, 0.308	Bone scanning		69
^{169}Yb	^{168}Yb$(n,\gamma)^{169}$Yb	32 days	EC	0.063, 0.110, 0.177, 0.198, 0.308		Enriched target	9
^{176m}Lu	^{175}Lu$(n,\gamma)^{176m}$Lu	3.7 hr	β^- (1.3)	0.088	Bone scanning		69
^{177}Lu	^{176}Lu$(n,\gamma)^{177}$Lu	6.7 days	β^- (0.497)	0.113, 0.208	Bone scanning		69

Appendix I (Continued)

Radio-isotope	Production methods	Half-life	Max. energy of major decay, MeV	Major photon energies, MeV	Some medical applications	Comments	References
^{191m}Ir	^{190}Os$(n,\gamma)^{191}$Os $\xrightarrow[15\text{ days}]{\beta^-}$ ^{191m}Ir	4.9 sec	IT	0.129		Isotope generator	
^{192}Ir	^{191}Ir$(n,\gamma)^{192}$Ir	74 days	β^- (0.67)	0.296, 0.308, 0.317, 0.468, 0.609	Tumor implants		18
^{194}Ir	^{193}Ir$(n,\gamma)^{194}$Ir	17.4 hr	β^- (2.24)	0.328			
^{198}Au	^{197}Au$(n,\gamma)^{198}$Au	2.7 days	β^- (0.962)	0.412	Aerosol lung scanning, liver function, tumor implants		20, 86
^{197}Hg	^{196}Hg$(n,\gamma)^{197}$Hg	65 hr	EC	0.077, 0.191	Kidney function	Preferred over ^{203}Hg, enriched target	8, 9, 18
^{203}Hg	^{202}Hg$(n,\gamma)^{203}$Hg	47 days	β^- (0.214)	0.279	Kidney function, brain scanning	Enriched target	8, 9, 18, 87
^{238}Pu	Neutron capture from ^{238}U	86 years	α (5.5)	U X rays	Power source (heart assist)	Needs to be free of ^{236}Pu, ^{17}O, ^{18}O	62
^{242}Am	Neutron capture from ^{238}U	16 hr	β^- (0.67)	Pu, Cm X rays	X-ray fluorescence, thyroid scans		65
^{252}Cf	Neutron capture from ^{238}U	2.6 years	α (6.1), SF	Fission neutrons	Tumor implants, neutron radiography		8, 88, 89

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AN IMPLANTABLE NUCLEAR-POWERED PHRENIC-NERVE SIMULATOR*

Preliminary development work has been done at NUMEC (Nuclear Materials and Equipment Corp.) on an implantable nuclear-powered diaphragm stimulator—phrenic-nerve simulator—to induce respiration where natural breathing is impaired. The initial experimental system, powered with a 2-W ^{238}Pu oxide source, is 108 cm^3 in volume and weighs 198 g. The system (Fig. 1) is contained in a hermetically sealed titanium case—about the size of the larger chemical-battery pacemakers but producing 20 times the electrical energy.

One prototype system has been constructed and tested. After 1 year of laboratory operation, the system was still performing satisfactorily. The results of the preliminary test indicate the feasibility of producing a radioisotope-powered implantable system in the milliwatt power range. (MG)

*Summary of a paper by T. F. Hursen, S. A. Kolenik, W. Johnson, S. Furman, and B. Parker, presented at the Interscience Energy Conversion Engineering Conference, Boston, Aug. 3–5, 1971, pp. 272–276 of Proceedings.

Fig. 1 First experimental model of nuclear-powered phrenic-nerve simulator.

METHOD FOR MEASURING MINERALIZATION IN LAKE SEDIMENTS*

There is no completely satisfactory method for in situ determination of the activity of microorganisms responsible for mineralization of organic matter on, for example, lake bottoms. A method, which, to date, has been tested only on sediment dredged from a lake bottom, was devised in which glucose–^{14}C is introduced. The rate at which $^{14}CO_2$ is released by the microorganisms indicates their metabolic rate and hence their mineralizing capability. Other labeled substrates might be used. Hopefully, further development will render the method applicable in situ. (MG)

*Abstract of article by M. J. Harrison, R. T. Wright, and R. Y. Morita, *Appl. Microbiol.*, **21**: 698-702 (1971).

In Vivo Tissue Analysis with ^{109}Cd

By Luther E. Preuss and Frank P. Bolin*

Supplementary Keywords: *source, gamma; comparison.*

Abstract: *A system has been developed for in vivo quantitative analysis of soft tissue. Preliminary work has indicated that ^{109}Cd is ideal for such an analysis, and use of a ^{109}Cd source has permitted determination of the percentage of fat in a given sample to within 2%. The attainment of this precision depends strongly on the initial assumptions regarding body composition. A composition of 77.4% O, 9.03% C, 10.0% H, 2.69% N, 0.31% K, 0.24% S, 0.19% P, 0.08% Na, and 0.08% Cl is considered to be representative of fat-free tissue, whereas 77% C, 11.5% O, and 11.5% H is the composition of choice for fat. Absorption experiments on assorted fats and lean meats support these assumptions. In addition, in vivo results have been obtained on humans and correlated with skinfold-thickness measurements.*

In the last 30 years there has been a great deal of interest in gross body composition. Numerous experiments have been performed utilizing a variety of techniques to determine one or more body variables—e.g., fat, bone mineral, lean body mass, or body water. The methods range from straightforward chemical analyses of cadavers to indirect systems such as specific gravity determination by underwater weighing. All of these systems have met with only limited success.

This report describes the development of a means of qualitatively determining the gross components of soft tissue—fat and fat-free. This particular method utilizes selective absorption of two monochromatic radiations and yields the percent fat content of the irradiated region. Advantages are accuracy, speed, and simplicity.

The system allows one to quickly determine the percent fat of a suitable area of the body, such as the triceps or calf muscle. Such determinations can conceivably be correlated with normal or diseased states of the human body and may be of great assistance to medical personnel in the diagnosis and management of illness.

THEORY

The exponential law of absorption can be written:

$$\ln (I_0/I) = \mu\rho x$$

where I_0 and I are the incident and transmitted intensities of a monoenergetic beam, ρ the density, x the path length, and μ the total mass-attenuation coefficient of the irradiated sample. By using two beams of different energies on the same sample, the ratio R may be established:

$$R = \frac{\ln (I_{01}/I_1)}{\ln (I_{02}/I_2)}$$

$$R = \frac{\mu_1 \rho x}{\mu_2 \rho x}$$

$$R = \frac{\mu_1}{\mu_2}$$

R, thus determined, is independent of density and thickness.

The percentage of fat can be deduced from R as follows: If the sample consists of two components A and B, then let the fractional part of each component be represented by F_A and F_B (where $F_A + F_B = 1$). It follows then that

$$R = \frac{\mu_1}{\mu_2}$$

$$R = \frac{F_A\,\mu_1^a + F_B\,\mu_1^b}{F_A\,\mu_2^a + F_B\,\mu_2^b}$$

If we assume that $\mu_2^a \simeq \mu_2^b$, as is the case with ^{109}Cd, then

$$R = \frac{F_A\,\mu_1^a + F_B\,\mu_1^b}{(F_A + F_B)\,\mu_2^a}$$

$$R = \frac{F_A\,\mu_1^a + F_B\,\mu_1^b}{\mu_2^a}$$

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$$R = F_A R_A + F_B R_B$$

$$R = F_A R_A + (1 - F_A) R_B$$

$$R = R_B + F_A (R_A - R_B)$$

Inverting,

$$F_A = \frac{R - R_B}{R_A - R_B} \qquad (1)$$

where R_A and R_B are the R values of the pure components A and B. If F_A is taken to be the fraction of fat in the sample, then a single determination of R will give the percent fat, since R_A and R_B can be found theoretically (Fig. 1).

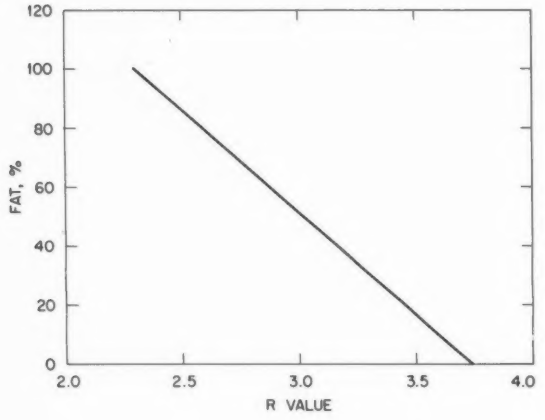

Fig. 1 Linear relationship between percent fat and R value of soft tissue.

The success of this technique depends on dividing soft tissue into two components: fat is taken to be 77% carbon, 11.5% oxygen, and 11.5% hydrogen by weight; fat-free tissue (FFT) has an assumed composition shown in Table 1.

X-ray machines and radionuclides have been considered as radiation sources for this technique. In addition to being less hazardous to both subject and user than X-ray generators, radioisotopic sources offer stable output, compactness, and monochromaticity; ^{109}Cd, ^{125}I, and ^{241}Am have been studied as possibilities.

Cadmium-109 offers several advantages over ^{241}Am and ^{125}I for this work. First, the intensity of the low-energy (22-keV) radiation from ^{109}Cd exceeds that of the 27-keV ^{125}I X ray when both are filtered

Table 1 Elemental Composition of FFT from the Literature,[1] Adjusted for 3% Fat Content

Element	Wt. %	μ 22	μ 88
O	77.4	0.670	0.162
H	10.0	0.366	0.301
C	9.03	0.368	0.157
N	2.69	0.475	0.158
K	0.31	8.10	0.279
S	0.24	4.85	0.232
P	0.19	4.00	0.209
Cl	0.08	5.95	0.256
Na	0.08	1.68	0.173
Mg	0.02	2.05	0.183

to 99.9% monochromaticity. Second, the difference between the mass absorption coefficients of fat and FFT is 50% greater with the 22-keV radiation than with the 27-keV photons; this indicates that ^{109}Cd is more sensitive to soft-tissue composition than is ^{125}I. Third, the half-life of ^{109}Cd (453 days) is much longer than that of ^{125}I (60 days). Finally, there is the obvious advantage of having a single source for both radiations.

The source currently used is 6 mCi of ^{109}Cd sealed in the conical tip of a Lucite cylinder. The cylinder is positioned inside a rectangular tungsten holder which can be rigidly attached to a frame so as to be immobile with respect to the detector aperture.

PROCEDURE

A block diagram of the equipment is shown in Fig. 2. (A linear amplifier has since been added.) The multichannel analyzer is used principally to monitor any changes in the gain of the system. The system is

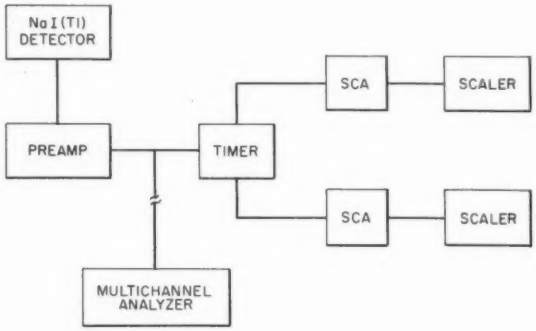

Fig. 2 Detection equipment used in the analysis of soft tissue.

capable of analyzing both in vitro and in vivo tissue specimens. In vitro samples are placed in hollow Lucite cylinders, and the ends are then sealed with a 1-mil sheet of saran and the cylinders interposed between the ^{109}Cd source and the detector and exposed to the beam for about 5 min. The intensities of the 22- and 88-keV radiations are taken from the scalers and are used to calculate the percentage of fat by means of Eq. 1.

For in vivo studies, a device has been constructed which will hold a human arm or leg in place while the area of the triceps muscle or calf muscle is being irradiated. A 10-min exposure of the triceps muscle results in a dosage of about 2 mrads to about 3 cm^3 of arm tissue, which is only about 1% of the dosage received from a single chest X ray.[2] Furthermore, the source holder and detector housing are mounted to a frame that is free to move along a metal track. A worm gear and reversible motor have been coupled to this frame, allowing one to scan across an area of tissue, thereby minimizing effects due to local inhomogeneities. The scanning system has an overall traverse of about 12.7 cm at a maximum rate of 1.4 cm per minute. The dosage, of course, is reduced by the scanning technique. The source strength makes it feasible to analyze tissue with thicknesses between 2 and 9 cm.

RESULTS

Experimental measurements on fat samples and FFT simulators have yielded R values that differ by less than 0.3% from those predicted on the basis of known sample composition and attenuation coefficients.[3] The theoretical R value for 100% fat is 2.30, and this can be compared with experimental values for various fatty substances as shown in Table 2.

Table 2 Experimental R Values for Lipid*

Fat	$R \pm 0.01$
Cod-liver oil	2.30
Corn oil	2.31
Olive oil	2.30
Rendered lard	2.31
Beef fat	2.30

*Dual-beam absorptiometric determinations on varied samples consisting of essentially 100% lipid result in R values which are in close correspondence; 2.30 is the theoretical value for 100% lipid.

In vitro studies were also carried out on several different meat samples. After being irradiated, a representative portion of each sample was selected and the fat content determined by ether extraction. Table 3 compares percent fat obtained chemically from various meat samples with that obtained by calculation of the R value. All R values listed are averages of at least five separate determinations, with deviations in all cases less than ± 0.02 in R.

Table 3 Muscle Sample Composition*

Sample	R value	% fat from R	% fat by analysis	Difference
Cattle	3.41	21.2	21.5	0.3
Cattle	3.38	22.8	22.9	0.1
Cattle	3.44	18.9	19.8	0.9
Cattle	3.40	21.9	20.6	1.3
Swine	3.42	20.1	21.2	1.1

*The percentage fat calculated from R measurements on assorted muscle samples is compared with that obtained by chemical analysis.

The initial study on humans involved the determination of the fat content of the triceps area on a small group of physically normal subjects. Measurements were carried out both by irradiating a fixed point and by scanning over an area. These values were correlated with skinfold measurements made with Lange calipers. Table 4 presents the percent fat from fixed-point and scan measurements as well as the skinfold values. The data are averaged for repeated measurements on the six subjects. Single-point values appeared to be unreliable, since a range of up to 6% variation was encountered. This can be attributed to the difficulty of exactly

Table 4 A Comparative Study on Human Subjects of Skinfold Thickness and Absorptiometric Determinations*

Subject	% fat (single point)	% fat (scan)	Skinfold thickness
1	9.3	6.3	0.5
2	12.0	11.3	1.1
3	15.4	16.4	1.0
4	18.4	23.0	1.5
5	30.5	31.6	2.8
6	47.2	43.3	3.1

*The correlation coefficient is 0.95 between the two methods of measurement.

repositioning the arm. The scan measurement was highly reproducible, however, having a maximum range of 0.5% in fat content. In addition, the percent fat determined by scan measurements correlated highly with skinfold-thickness measurements, the correlation coefficient being 0.95.

Future plans include in vivo testing on healthy volunteers and, later, on patients with some relevant type of disorder (e.g., obesity).[4] Points for study will be both long-term reproducibility of the scan measurements and comparison with fat content determined at other locations of the body, such as the calf.

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Section

VI

Isotopes and Radiation Technology

Safety

Radioactive Sources for Safeguards and Materials Management

By H. O. Menlove, R. A. Forster, and J. E. Foley*

Supplementary Keywords: *safety; industry, nuclear; source, neutron; chemistry, analytical; activation analysis; comparison;* ^{124}Sb; ^{88}Y; ^{226}Ra; ^{238}Pu; ^{252}Cf.

Abstract: *Because of the simplicity and reliability of radioactive neutron sources, they are expected to play an important role for in-plant measurements of fissionable materials and quality control of reactor fuel components. A given nondestructive assay problem requires optimization of the neutron source, moderator, and detector, and strong calculational capability is needed to make preliminary evaluations on the combinations of these three sets of variables.*

Neutron interrogation has been used extensively during the past few years for the nondestructive assay of fissionable materials. The usefulness of this technique is due primarily to the uniqueness of the neutron-fission reaction to the fissionable material of interest. In addition, the neutrons have good penetrability and the neutron-fission cross sections give useful isotopic signatures. Either accelerator or radioisotopic neutrons could be used. However, although accelerator neutron sources offer large yields and source modulation flexibility (i.e., pulsing), they usually require a substantial amount of initial investment and technical support. On the other hand, radioactive neutron sources are reliable, simple, and less costly and are well suited for in-plant measurements and quality control of reactor fuel components as well as for some categories of scrap-material and process-stream measurements.

*University of California, Los Alamos Scientific Laboratory, Los Alamos, N. Mex. 87544. This paper in its complete form was presented at the 12th Institute of Nuclear Materials Management Meeting, Palm Beach, Fla., June 29–July 1, 1971.

In considering the application of radioactive sources to a given measurement problem, three primary variables should be considered: (1) the type of neutron source, (2) the moderator and shield for the source, and (3) the detector to count the induced neutrons and/or gamma rays. A good assay system is one that combines the three components in an optimum manner for the given problem.

SOURCE SPECTRUM AND MODERATOR CONSIDERATIONS

Radioactive neutron sources under investigation at LASL include ^{124}Sb–Be(γ,n), ^{88}Y–Be(γ,n), ^{226}Ra–Be(γ,n), and ^{238}Pu–Li(γ,n) (which have neutron energies primarily below the fission thresholds of ^{238}U and ^{232}Th), and ^{252}Cf (which emits higher energy neutrons). Since the ^{252}Cf source fissions spontaneously to emit neutrons with both sub- and superthreshold components, it can be used to give both fissile and fertile isotopic measurements using the two-spectrum irradiation technique.[1] Some of the characteristics of these and other sources for nuclear safeguards applications have been summarized previously.[2]

In the application of radioactive sources, it is usually necessary to surround the irradiation system with neutron and gamma shielding to protect personnel and detectors from the source radiation. This shielding material significantly lowers the average energy of the irradiation neutrons, with corresponding decreases in neutron penetrability. To understand and predict the influence of various moderator materials on the measurement system, a substantial calculational program is

in progress to optimize neutron-irradiation configurations for given assay problems. Some of these calculated results have been reported previously.[3]

A composite moderator assembly (Fig. 1) was used with a 0.7-mg ^{252}Cf neutron source to assay the fissile content of several UHTREX (Ultra High Temperature Reactor Experiment) fuel rods. These rods are 6 in. long, have an outer diameter of about 1 in., and are made of enriched-uranium (93% ^{235}U) beads coated with graphite. The rods had been irradiated for 107 MWd, giving a measured dose rate of ~1000 R/hr at contact when the assays were made. Source neutrons for the interrogation were obtained from a ^{252}Cf source located in the center of the assembly, and the fuel rods were passed through a hole in the hydrocarbon sample port. A small ^{238}U spiral fission chamber was placed in the detector hole to count the prompt neutrons from induced fissions in the fuel rod.

For measurements on hot fuel elements, it is necessary to either use a neutron detector that is insensitive to the high gamma-radiation levels or shield the detector with a substantial amount of heavy material. Ionization fission chambers are very insensitive to gamma radiation, and it was possible to place the ^{238}U fission chamber next to the hot fuel elements without any increase in the background rate.

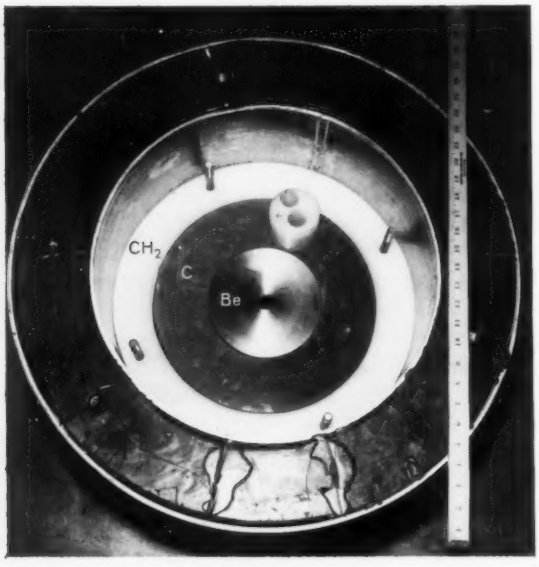

Fig. 1 Interior of moderating assembly showing the central hole, which normally contains the ^{252}Cf or (γ,n) neutron source, and the sample–detector part at the intermediate radial position.

The results of the prompt-neutron assay of hot UHTREX rods are given in Table 1. A calibration curve (Fig. 2) was obtained by scanning three cold fuel rods, with known uranium loadings, with the interrogation assembly. The nonlinearity in the curve is due to thermal-neutron absorption in the fuel material.

Table 1 Active Assay of Hot UHTREX Fuel Rods Using a ^{252}Cf Neutron Source

UHTREX rod No.	Nominal loading,* g of U	Prompt-neutron assay, g of U
603	6.6	6.55
4238	7.6	7.59
4523	8.9	8.75
5115	10.5	10.40

*Nominal loadings were measured during fabrication using a γ-ray scan.

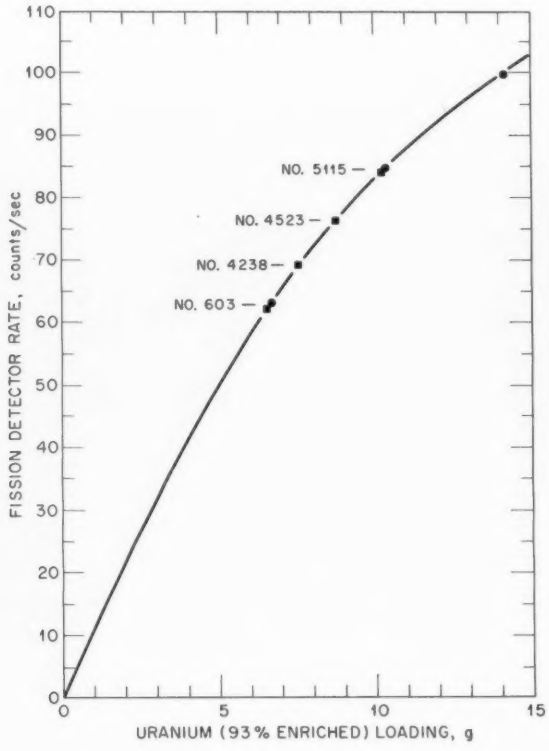

Fig. 2 Detector response vs. uranium loading calibration curve for the assay of UHTREX fuel rods. The serial numbers identify the unknown hot fuel rods.

ASSAY SYSTEMS AND APPLICATIONS

^{252}Cf Fuel-Rod Assay System

The neutron-transport calculations described in Ref. 3 were used to optimize the design of a ^{252}Cf moderator system for assaying the fissile content of uranium or plutonium fuel rods and other small samples. Moderated neutrons from the ^{252}Cf source interrogate the fuel material, and prompt neutrons from the induced fission reactions are counted, using energy-biased helium-gas recoil detectors. Such a system is limited to material with low fissile-content density because of self-absorption of thermal neutrons in the fuel.

The ^{252}Cf fuel-rod assay system (Fig. 3) includes a fully engineered six-channel rod-handling mechanism and data acquisition, reduction, and printout equipment. A programmed calculator and interface unit make it possible to reduce the raw data, rapidly print out the assay value, and automate the measurement procedure for in-plant applications. Since the high-energy neutrons from the ^{252}Cf source which reach the detector have essentially the same spectrum as the signal neutrons from fissions in the sample, this constant background rate has been utilized in a feedback loop to automatically stabilize the response of the system.

The assay system has been used for both UO_2 and UO_2–PuO_2 fuel rods. The net counting rate for a typical boiling-water reactor (BWR)-type fuel rod (2.34% enriched in ^{235}U) was measured to be >1200 counts/sec for a 100-μg ^{252}Cf source. The measured

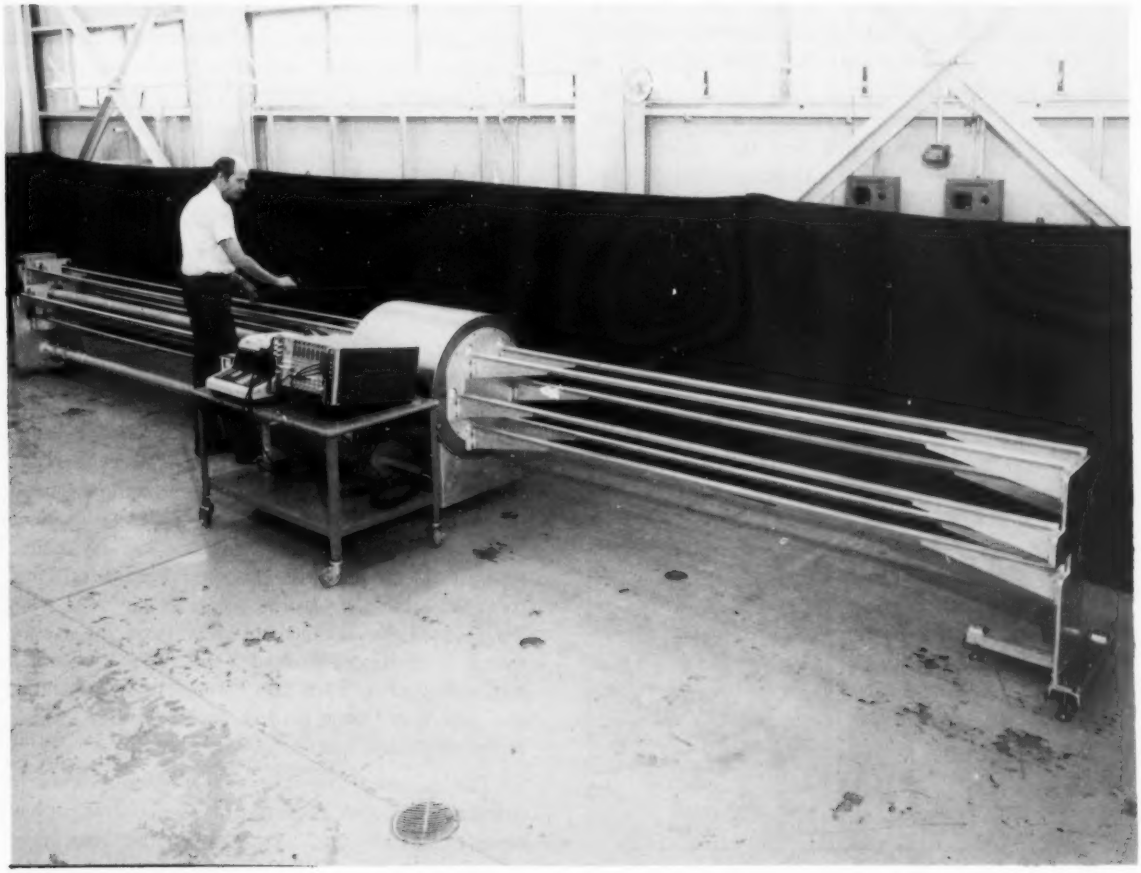

Fig. 3 Complete ^{252}Cf fuel-rod assay system with six-channel rod-handling mechanism and automated data acquisition, reduction, and printout. Assay accuracy <1% for fissile content of power-reactor fuel rods; assay time ~30 sec for six simultaneous channels.

counting rate of a plutonium fuel rod with a loading of 0.5 g of plutonium per inch was ~1400 counts/sec. Neither uranium nor plutonium fuel rods give a significant background in the detectors when the ^{252}Cf source is removed.

Delayed Gamma Rays from Fission Using Neutron Interrogation

The characteristics of delayed-gamma-ray signatures for fissionable-material assay have been described previously.[4] Although delayed gamma rays have a relatively large yield per fission and have conveniently long decay half-lives, the process of delayed gamma emission following neutron interrogation is common to many matrix and container materials, and therefore considerable care must be exercised in interpreting the results.

The ^{252}Cf fuel-rod assay system shown in Fig. 3 has been modified to measure the delayed gamma rays as well as the prompt neutrons from fission. The prompt neutrons are detected with ^{4}He-recoil counters. In addition, a gamma detector with appropriate lead shielding was positioned near the fuel-rod exit channel in the ^{252}Cf shield. Approximately 5 in. of lead was placed between the ^{252}Cf assembly and a 5- by 5-in. NaI(Tl) gamma detector to shield the detector from the neutron-source gamma rays. For the measurements, the fuel rod was pushed by the automatic rod driver through the ^{252}Cf neutron interrogation region, where the induced fission neutrons were counted, and then through a 2-in.-diameter hole in the NaI detector where the delayed gamma rays were counted. The bottom portion of Fig. 4 shows the passive gamma background from a BWR-type fuel rod (2.34% enriched in ^{235}U), followed by the gamma response of the rod as it is pushed through the irradiation and counting regions at a uniform speed (~5 ft/min). To study background levels, signal rates, and decay rates, the curve is separated into the four regions: (1) passive rod background plus ^{252}Cf source background; (2) ^{252}Cf source background; (3) induced delayed gamma signal plus backgrounds; and (4) delayed gamma time decay of the rod after the rod motion was stopped while the rod was in the NaI detector. In addition to the BWR-type enriched-uranium fuel rod, a similar rod containing 2.4% plutonium was investigated with the NaI detector. The results are shown in the top portion of Fig. 4. In both cases shown in Fig. 4, the detector was biased to count gamma rays with energies greater than 1.5 MeV; even for these hard gammas, the net signal rate in both cases is greater than 5000 counts/sec.

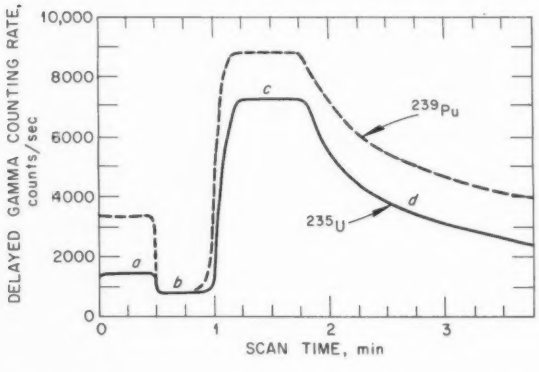

Fig. 4 Gamma response of a BWR-type fuel rod and a similar rod with plutonium replacing ^{235}U.

These delayed gamma-ray measurements, together with calculational studies, are being used to develop an optimized gamma-ray detection system for pellet-to-pellet scanning of power-reactor fuel rods. The rod-handling mechanism of the present ^{252}Cf fuel-rod assay system will be modified to accommodate gamma detection so that complete pellet-to-pellet scan information can be obtained when desired.

Random-Source Neutron Interrogation Using Coincidence Counting

Neutron coincidence counters have been used extensively during the past few years for the passive assay of ^{240}Pu and more recently for ^{238}U. Although ^{235}U has an insignificant spontaneous fission rate, it has been found that the neutron-coincidence technique can be extended to ^{235}U by using a random neutron source to induce fissions in the ^{235}U. The coincidence logic unit of the neutron detector can be used to separate the counts of the random interrogation source from the subsequent fission events in the ^{235}U, which are induced by the neutron flux from the random source. A neutron source such as ^{210}Po–Li would be satisfactory since it has no spontaneous fission component but does have a neutron spectrum that is almost completely subthreshold.

The required assay time, the optimum detector characteristics, and interrogation source strength have been calculated, and a random-source coincidence assay system (Fig. 5) is under construction at Los Alamos Scientific Laboratory to evaluate the technique. The optimum choice among various types of detectors—including solid and liquid scintillators—

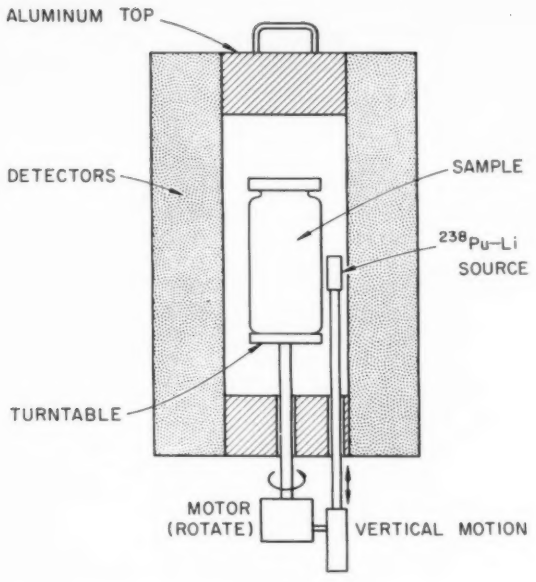

Fig. 5 Assay system for determining ^{235}U content using new random-source interrogation technique.

and the closely related problems of detector-associated matrix effects are being investigated in detail, using appropriate computational programs. (MG)

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Section

VII

Isotopes and Radiation Technology

Isotopes Around the World

Recent Descriptions of Irradiators Abroad

Supplementary Keywords: *facility description; source, gamma;* ^{60}Co.

USSR

Two Soviet irradiation facilities were described in a recent issue of the USSR atomic energy journal—one for continuous radiation processing of either liquid or gaseous reactants and the other for use in laboratory studies.[1,2]

There are two models of the chemical processing facility—the RKhUND-20000 (Fig. 1) and Gammatok—which is of the "squirrel cage" type. Both use ^{60}Co as the radiation source. The 20-kCi source of the RKhUND-20000 provides a dose rate of 220 R/sec in the 9-liter reaction chamber. The 80 cylindrical pellets of ^{60}Co (250 Ci each) are contained in 20 tubes, which surround the reaction chamber. Inlet and outlet tubes to the chamber and a stirring mechanism are provided. The Gammatok has a total activity ot 56 kCi (80 pellets, 700 Ci each, distributed in 16 tubes) and provides a dose rate of 260 R/sec in its 35-liter irradiation chamber.

The RMKh-γ-25M is an irradiator that may be located in an ordinary laboratory for use in research. It is a modified version of the RMKh-γ-100, a facility widely used in the USSR.[3] The irradiation chamber has a volume of 1.2 liters and capacity for a 16.2-kCi ^{60}Co source, which would deliver a dose rate of 2×10^6 R/hr. Provision is made for irradiation under pressure up to 100 atm and at constant temperature. Among the uses for which it is suited are manufacture of vaccines by inactivation of the living organisms and sterilization of blood serum and transplants.

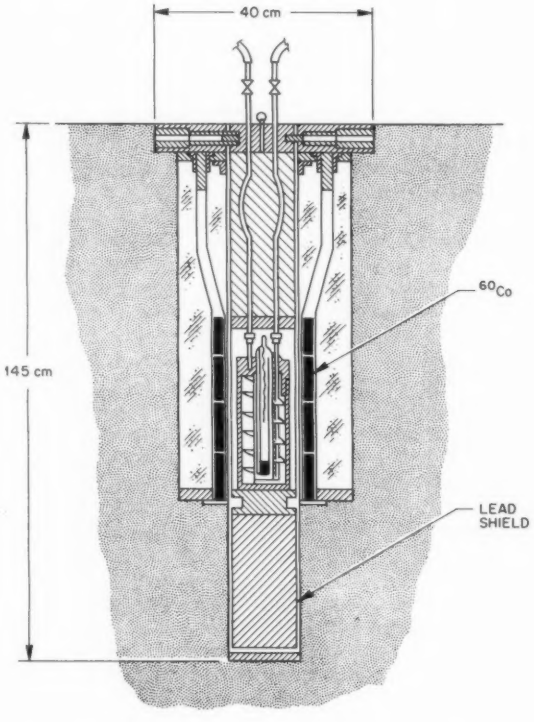

Fig. 1 Vertical section of USSR RKhUND-20000 irradiation facility.

POLAND

An irradiation facility (Fig. 2) for use in physico-chemical investigations has been installed at the Institute of Nuclear Research, Warsaw-Žerań.[4] The 960-Ci

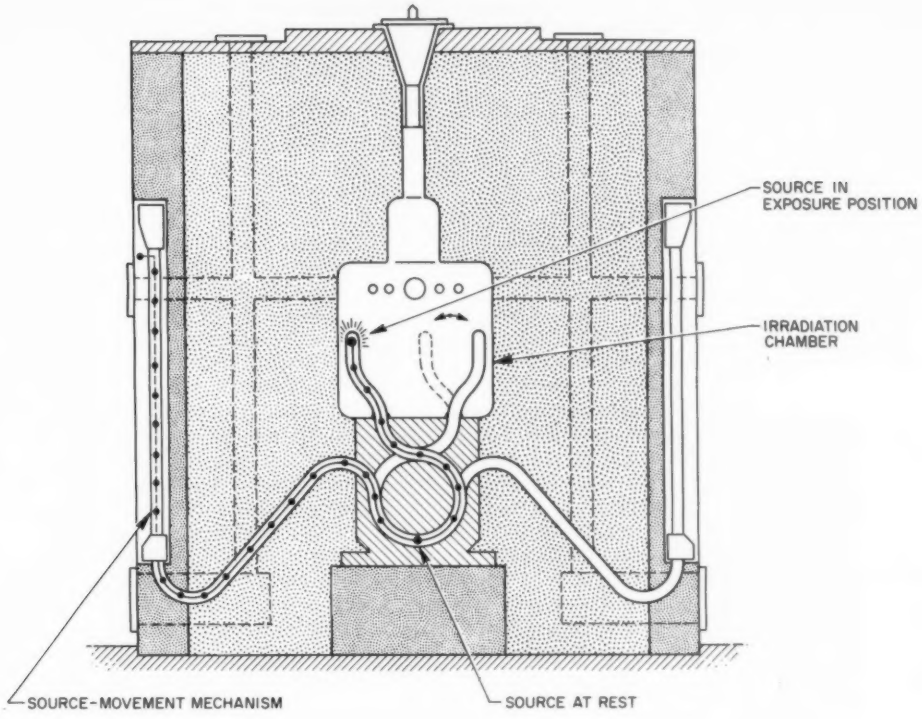

Fig. 2 Vertical section of Polish-type 180-LG irradiator.

^{60}Co source, obtained from the USSR, is made up of eight pellets, 120 Ci each. The sources are located in eight tubes, which are bent and can be rotated to change the dose rate at the center of the 125-liter cubical irradiation chamber from 3.0×10^4 to 6.5×10^5 rads/hr. Provision is made for introducing gases and liquids to the chamber and for supplying electricity so that studies can be made in flow systems or at high temperatures. The source pellets are moved by manipulating a lever in the wall which connects with the source subassemblies. The shielding is 30 tons of 0.5- to 5-mm shot.

FRANCE

In the past 2 years, a French company, Conservatome,[5] has designed and installed four irradiators with ^{137}Cs. "Lisa 2," for irradiating small animals, has a 40-liter irradiation chamber, and its 2-kCi source delivers a dose rate of 40 to 200 rads/min. A "Lisa 3" model, which is suitable for experimental processing of food, was installed on the Ivory Coast. Its irradiation chamber is 70 liters in volume and can accommodate an 80-kCi source. "Irpa 1" has a 2-kCi source and a geometry similar to that used for teletherapy. Conservatome also has supplied a 2-kCi unit for extracorporeal irradiation of blood to the Grenoble Hospital.

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BRITISH GAMMA-IRRADIATION FACILITY*

A gamma-sterilization plant at Reading, England, is the only company in the United Kingdom whose sole activity is the gamma irradiation of manufacturers' products. The $3.6 million plant, built by H. S. Marsh Ltd., measures 44 by 41 by 17 ft high and is housed in a warehouse building with a floor area of 15 thousand ft^2. The irradiation cell, in the center of the plant, has 5.5-ft-thick concrete walls and a 10-ton electrically operated door. The 350-kCi ^{60}Co source is in the center of the cell, which incorporates a dry pit for safe storage. The source consists of a layer of tubes about 2 ft long and 1.5 in. diameter. Each tube is filled with helium, and the closure is argon-arc welded. Twenty-four cobalt rods (about 6 by 0.5 in.), each in a helium-filled stainless-steel tube, are located in four groups of six rods each inside the larger tubes.

Packages to be irradiated are loaded onto an input conveyor (Fig. 1) and then, by completely automated control, transferred to carriers suspended vertically from an overhead monorail. Box sizes up

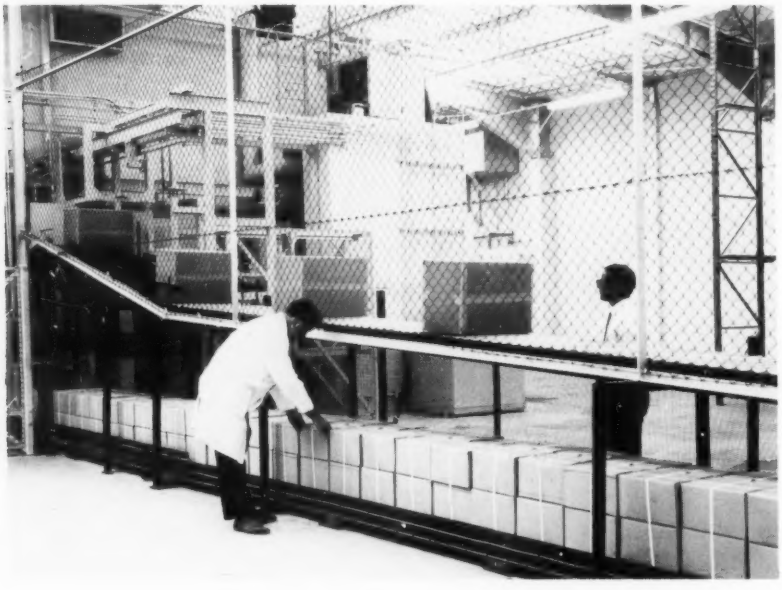

Fig. 1 Packages on input conveyor, ready to be started on the irradiation process.

to a maximum of 23.5 by 17 by 13.75 in. can be handled. After a 12- to 48-hr residence in the cell, during which the packages are transported around the source five times at different heights in the carriers to ensure uniform dosage, the carriers are unloaded and the packages are collected by the customer. The U. K. standard dose of 2.5 Mrads is given unless the customer specifies a different dose, in which case a secondary conveyor, installed for this specific purpose, is used. Labels that change color upon irradiation are attached to every box as a check that the process has been applied, and the radiation dose is measured by spectrophotometric observation of the change in the optical characteristics of plastic strips attached to a few of the packages before irradiation.

During its first year of operation, the plant is processing disposable medical products, but in the future other products will be treated, including wood–plastic composites and some foods, which will be pasteurized experimentally. (MG)

*Operated by Gamma Radiation Services Limited, Marcus Close, Dee Road, Tilehurst, Reading, Berks. The material for this item was supplied by the company.

USSR HIGH-INTENSITY GAMMA FACILITY*

Summarized by Martha Gerrard

A gamma-irradiation facility, the UGU-200, with a total source activity of 400 kCi, was started up in 1968 at the Byelorussian Academy of Sciences' Institute of Nuclear Energy. It is designed for experimental and semi-industrial-scale work on uses of gamma radiation.

The source consists of two units of about 200 kCi each. Each unit is made up of 50 cylindrical stainless-steel cartridges containing six doubly encapsulated (stainless steel, 81.5 mm long, 11 mm diameter) standard 700-kCi ^{60}Co sources, for a total of 4200 kCi per cartridge. The source construction is such that in only 4 or 5 days the parts can be reassembled into a plane-parallel source, cylindrical sources of different diameters, or other geometrical shapes.

When not in use, the source is stored inside a reinforced concrete block on the bottom of a concrete-shielded pool (6.4 m deep, 6 m diameter) of desalted water (Fig. 1). The source is mounted on a

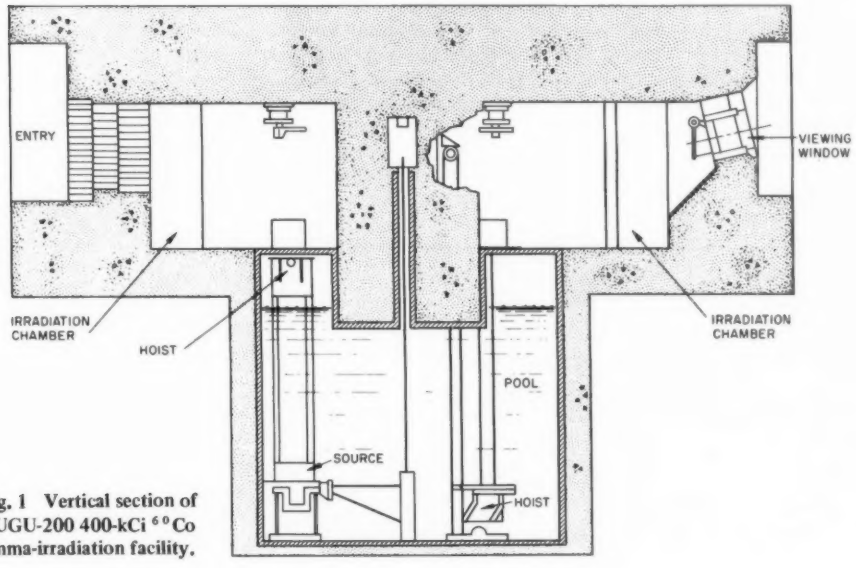

Fig. 1 Vertical section of UGU-200 400-kCi ^{60}Co gamma-irradiation facility.

removable platform, which is attached to a vertical support that can turn as much as 240° and thus readily transfer the source, under water, to another part of the pool. The platform consists of two spacer disks clamped together and may be moved from the rotating arm to the hoist that lifts the source into the irradiation position.

The upper part of the cylindrical facility is divided into three sections. One section is outside the shield and is for loading and maintenance. The other two, which have a volume of 35 m^3, are for sample irradiation. The shielding on these is sufficient for use of a 210-kCi source, and they may be used simultaneously or one may be in use while the other is being prepared for an irradiation. Thirty-two tubes through the walls allow communication between the outside and the chamber when necessary, and five vertical channels in the roof connect the irradiation chambers with the workrooms. Various types of measurements can be made while the object is being irradiated, and viewing windows allow observation during the process. Inside the chambers are mounting stages, 1.4 by 1.3 m, for large objects.

The irradiation field can be varied from 1 to thousands of R/sec.

*Summary of article by A. K. Krasin, I. I. Salamatov, V. A. Borisevich, A. V. Polyushits, A. R. Latsuk, and A. N. Rozdyalovskii, *Atomnaya Energiya (USSR)*, **31**: 205-207 (1971) (in Russian).

AUSTRALIAN IRRADIATION FACILITY FOR AGRICULTURAL RESEARCH*

Supplementary Keywords: *facility description; source, gamma;* ^{60}Co.

The irradiation facility of the C.S.R. Research Laboratories in Roseville, New South Wales, was designed and built by Unisearch Ltd., Sydney. In January 1969, the facility was loaded with 1675 Ci of ^{60}Co. The source—two pencils of cobalt, each 15.6 cm long—is stored in a 4.5-ton lead-filled steel casing, fabricated in such a manner that the operators are protected from the source at all times. The facility is 0.84 m wide and 1.5 m high. This type of unit is more economical to construct than are shielded rooms, where the source is stored in a deep well of water when not in operation.

The vertically mounted cobalt pencils are located 20 cm from the central axes of two cylindrical irradiation chambers. The chambers are 15 cm high by 8.75 cm diameter and are raised and lowered pneumatically. In their lowered position they are exposed to the ^{60}Co source, and in their raised position they are accessible to the operator. Removable specimen containers fit neatly into the irradiation chambers.

To equalize exposure within a chamber, the containers are rotated at 20 rev/min by $^{1}/_{16}$-hp motors. The duration of an exposure is set on a time switch, which operates the mechanism to raise the chamber out of the radiation field at a predetermined time. During irradiation, compressed air, cooled by passage through refrigerant coils, can flow through the complete irradiation compartment. Provided that the organisms being irradiated are held in open-mesh containers, it is also possible to regulate their gaseous environment. (MG)

*Abstract of article by P. B Hutchinson and A. W. Osborn, *Atomic Energy in Australia*, **14**(2): 14-17 (April 1971).

Section

VIII

Isotopes and Radiation Technology

Miscellaneous

Availability of Isotopes and Services

^{252}Cf AVAILABLE AS SOLID WIRE

The Atomic Energy Commission is offering limited, developmental quantities of ^{252}Cf in solid wire form for sale. This new form is expected to encourage broader industrial participation in californium encapsulation since wet chemistry operations will not be necessary when the material is removed from the shipping container. The product is the result of a development effort by the USAEC's Savannah River Laboratory in South Carolina, following industry's request for a more readily usable form of californium. Initial orders will be accepted from U. S. customers only, because of the limited amount available. Sales of californium oxide and oxysulfate will continue to fill requests from both foreign and domestic customers unable to obtain the early supplies of solid wire, probably for about 6 months or longer if industry interest warrants. The price of the wire, as for the other forms, will be $10 per microgram of ^{252}Cf and a $1600 handling packaging charge F.O.B. Savannah River Plant.

Additional information on procedures for purchasing this new form including material specifications, descriptions of the shipping packaging, and sales order forms may be obtained upon request from the Manager, Savannah River Operations Office, U. S. Atomic Energy Commission, P. O. Box A, Aiken, S. C. 29801.

NEW ^{99m}Tc GENERATOR

The "Ultra-TechneKow," a new generator of sterile ^{99m}Tc, is being offered by Mallinckrodt Chemical Works (St. Louis, Mo. 63160). The instrument features shielding that minimizes operator exposure and an assaying attachment. The generator is supplied with 50, 100, 200, or 300 mCi of the ^{99}Mo parent.

NUCLEAR MOISTURE GAGE

A bulk moisture gage that contains both a neutron and a gamma source is announced by The Ohmart Corporation (4241 Allendorf Drive, Cincinnati, Ohio 45209). The fast-neutron source and the ^{137}Cs gamma source are housed in the lower arm of a "C" frame and detectors in the upper. The C frame can be mounted on a conveyor belt at any time without interruption of operation. Moisture measurement precisions of ±0.5% for sinter mix and taconite and of ±1% for wood chips are claimed.

SOILTEST MARKETS NUCLEAR MOISTURE–DENSITY METER

A portable (18 lb), simple, all-weather, high-speed, nondestructive meter for use in determining moisture and density of soils, stabilized soils, and aggregates and density of asphalt mixtures is being marketed by Soiltest, Inc. (2205 Lee St., Evanston, Ill. 60202). It can be used on dam cores, earth dams, levees, embankments, and subbases for roads as well as on highways and airport pavements and around footings. The meter (NIC-5) has three systems that operate simultaneously but independently—a thermal-neutron system with a conventional boron trifluoride detector for moisture determination and two gamma-backscatter systems for density determination. Power is supplied by rechargeable nickel–cadmium batteries, and operating time without recharging is about 40 hr. The standard radiation source is ^{226}Ra–Be, but ^{137}Cs and ^{241}Am–Be may be used.

AUTOMATED EDX SYSTEM MARKETED BY QANTA/METRIX

A completely automated energy-dispersive X-ray (EDX) analysis system that combines an advanced solid-state EDX spectrometer with a preprogrammed minicomputer is offered by Qanta/Metrix, San Carlos, Calif. The system is controlled completely by push buttons and can be used by personnel with no data-processing knowledge. Qualitative or semiquantitative analysis usually requires less than 1 min, and accurate quantitative analysis usually takes only a few minutes. Three models are offered for research, the main differences being the programs available for analysis. A model with "go/no-go" controls for process monitoring could easily be designed. The basic system uses an X-ray machine, but a radioisotopic X-ray source is an available option and is being used in a system installed at a TVA laboratory at Muscle Shoals, Ala.

BRITISH X-RAY-FLUORESCENCE ANALYZER

An X-ray-fluorescence analyzer developed by Applied Research Laboratories Ltd. (Wingate Road, Luton, Bedfordshire, England) is claimed to have a sensitivity, in a light matrix, of from 100 ppM for silicon to 10 ppM for heavy elements. It uses either a radioisotope or an X-ray tube as radiation source and can make unattended, automatic routine analyses. The instrument was described by A. L. Gray at the Denver (Colo.) X-Ray Conference, August 1971.

"INSTANT" INDUSTRIAL RADIOGRAPHS

A system for rapid radiography, which consists of paper, screens, processor, and chemicals, is available from Kodak. The "Industrex" system is usable with ^{192}Ir and ^{60}Co radiation sources as well as with high- and low-voltage X-ray tubes and is claimed to be convenient, efficient, and economical as well as rapid.

MINIATURE RADIOISOTOPE-POWERED ENERGY SOURCE

Gulf General Atomic (P. O. Box 608, San Diego, Calif. 92112) announces the availability of the British-developed miniature radioisotope-powered energy source for use in cardiac pacemakers and elsewhere [*Isotopes and Radiation Technology*, 7(2): 194-196 (Winter 1969–1970)]. Gulf has been licensed to manufacture the ^{238}Pu-powered unit in the United States and to sell it in the United States and abroad.

ATOM AND ENVIRONMENT FILM AVAILABLE

"The Atom and the Environment," a 22-min color motion picture, describes numerous ways in which nuclides can be used in environmental studies—study of the growth of plants and trees and the movement of elements through grasses, shrubs, and trees; measurement of a flowing stream's ability to purify itself after waste is dumped into it and of conversion of CO_2 to O_2; investigation of the absorption by plants of pollutants without interference with photosynthesis; and the production, by neutron activation analysis, of "atomic fingerprints" that identify Hg, Cu, Zn, and As.

The film is designed for general audiences and may be obtained free of charge, except for return postage, from the Motion Picture Film Library, Oak Ridge Associated Universities, P. O. Box 117, Oak Ridge, Tenn. 37830.

AEC Activities

AEC MEETING WITH POTENTIAL ^{252}Cf SYSTEMS MANUFACTURERS

On Nov. 10, 1971, the USAEC sponsored a meeting in Augusta, Ga., with representatives from private industry on the use of ^{252}Cf in the areas of process control, nondestructive testing, nuclear materials safeguards, and neutron activation analysis. These manufacturers of devices, instruments, and hardware that could be used in conjunction with ^{252}Cf neutron sources had expressed interest in discussions of the characteristics and availability of ^{252}Cf and its potential uses. Several systems using ^{252}Cf which have already been developed in the AEC's Market Evaluation Program were also discussed. The current supply and demand picture for the radioisotope was reviewed, and information on licensing, foreign interest, and the AEC's loan and sales programs was presented. The

proceedings will not be published, but the meeting will be reported in an early issue of *Californium-252 Progress*, available from the Manager, Savannah River Operations Office, P. O. Box A, Aiken, S. C. 29801 [telephone (803) 824-6331, extension 2252].

U. S. TO DONATE ^{252}Cf SOURCES TO IAEA

Needles containing small amounts of ^{252}Cf are being given by the USAEC to the International Atomic Energy Agency for distribution to foreign universities in member states. The needles, fabricated at the AEC's Savannah River Plant in South Carolina, are now on loan to foreign medical institutions for cancer therapy studies. They will be given away, probably in about 18 months, when their activity has decayed to a level too low for medical studies but not too low for reactor experiments, dosimetry studies, instrument calibration, production of other short-lived isotopes, and some neutron activation analysis. The isotope is also available for sale to both foreign and domestic customers at a price of $10 per microgram, plus handling.

GENEVA FILMS ON ISOTOPES NOW AVAILABLE

Seven color motion pictures on isotope uses, produced especially for the Fourth United Nations International Conference on the Peaceful Uses of Atomic Energy in Geneva, Switzerland, September 1971, are now available for loan to the public from the Atomic Energy Commission. The films are available free of charge, except for return postage, from the Motion Picture Film Library, Oak Ridge Associated Universities, P. O. Box 117, Oak Ridge, Tenn. 37830. They are:

Doorway to Diagnosis (28 min): covers progress in development of instrumentation, developed under AEC sponsorship, for disease detection.

Isotopes in Environmental Control (14 min): illustrates use of radioactive atoms to help man preserve and restore his environment.

Nuclear Innovations in Process Control (17 min): depicts the versatility and sophistication of nuclear methods now available for control of industrial processes and for nondestructive testing.

Radiation Processing: A New Industry (14 min): gives examples of how the use of radiation for industrial processing in the United States has increased in recent years.

The Radioisotope-Powered Cardiac Pacemaker (21 min): depicts the fabrication and lifetime testing of the nuclear-powered cardiac pacemaker and shows the implantation of the device in a dog.

Short-Lived Radioisotopes in Nuclear Medicine (27 min): describes refinements in nuclear medicine which offer the physician a better diagnostic picture, faster, and with the lowest radiation burden to the patient.

Space and the Atom (27 min): reports past successes of nuclear energy in space, such as radioisotopic generators on recent Apollo lunar flights, and outlines efforts to develop a nuclear rocket engine.

FIRST ISOTOPIC PACEMAKER LICENSE

The first AEC license covering an isotopic pacemaker implanted in a human was issued on Sept. 20, 1971. Madame J. Labatut, Agence Photographique, Paris, was granted an 8-week license for possession of 200 mg of plutonium as a sealed source contained in an implanted Medtronic isotopic pacemaker, Laurens-Alcatel Model 9000. Madame Labatut, who had the pacemaker implanted in 1970, sought the license to cover a stay in the United States. She wears a gold identification bracelet that contains emergency notification information in both French and English.

General

CANADIANS INTERESTED IN WOOD–PLASTIC COMPOSITE FLOORING

Three Canadian companies—DuPont Development Division, Barwood of Canada, and Northern Flooring (Canadian agent for Arco Chemical Company's "Perma Grain")—are involved with wood–plastic composite (WPC) flooring in which the impregnating monomer is polymerized with gamma radiation. Atomic Energy of Canada Limited, the world's leading supplier of ^{60}Co

process units, has two process irradiators capable of polymerizing methyl methacrylate in impregnated wood. In one they have been doing irradiations for Barwood of Canada, and the other has been leased by Radiation Technology (Canada) Ltd. The installed cost for this flooring is about $1.70 per square foot at present, but a lower cost would help to increase acceptance. Large, i.e., 10^6 ft^2/year, plants would be more efficient and thus would reduce costs. Unfortunately, Canadian demand for WPC flooring is much less than this amount. Nevertheless, with an annual usage of $>10^8$ ft^2 of wood flooring, the WPC may eventually take as much as 10% of the overall market.

ISOTOPIC HEATERS TO PROTECT JUPITER MISSION EQUIPMENT

Nuclear-fueled heaters similar to those placed on the moon in the 1969 Apollo 11 flights will be used to prevent low-temperature damage to "Pioneer" spacecraft equipment during fly-by missions to Jupiter scheduled for launch in 1972 and 1973. The Atomic Energy Commission, which developed the isotopic heater technology, has already delivered 12 flight-qualified units to the National Aeronautics and Space Administration for the 1972 mission, with units for the second mission scheduled for delivery in March 1972. The Pioneer units, like the Apollo 11 units left on the moon, are fueled with ^{238}Pu but weigh less than 2 oz (vs. 2 lb for the moon units). The self-contained 1-W heaters—2 in. long and 1 in. in diameter—are capable of extended continuous operation. The unit is tested to withstand any credible temperature or accident without leaking, and a rocket flight test has confirmed that the heaters could withstand the extreme conditions of reentry into Earth's atmosphere without capsule rupture.

FRENCH ^{90}Sr-BASED GENERATOR FOR POLAR USE

A 24-V, 3-W generator, the Isotaf-1, is being developed by Ateliers de Chantiers de Bretagne (ACB) in cooperation with Alcatel and the Commissariat à l'Energie Atomique for French Polar use. The <250-kg unit is designed to operate for several years at −40°C. The Isotaf generators are powered with ^{90}Sr.

CARDIAC PACEMAKERS ABROAD

The West German Federal Ministry for Science and Technology has appropriated DM 14 million ($425,000) for the development of artificial hearts and lungs, with AEG-Telefunken, Siemens, and Messerschmitt-Bölkow and the surgical clinic of the University Hospital of Berlin-Westend collaborating. Within 2 years, the medical specialists and engineers hope to construct a two-chamber blood pump, simulating a heart.

A market survey made in France indicates that, in France alone, 500 cardiac pacemakers powered by ^{238}Pu will be needed by mid-1972, with this number reaching 5000 by the end of 1974. French facilities for ^{238}Pu production will be expanded, but several kilograms of ^{237}Np, from which ^{238}Pu is produced, will also be purchased abroad to meet the sudden increase in demand.

Two units of Japan's first nuclear pacemaker, developed by Daini Seikosha Co. Ltd. and Tokyo Womens' Medical College Hospital for implantation in dogs, are to be constructed for extensive testing, with 100 or more units expected to be produced for human implants within 5 years. The cylindrical 10-mm-high by 12-mm-diameter pacemaker has a theoretical output of 6 V. It weighs about 50 g and is powered with a 15-g Betacel (^{147}Pm) from Donald W. Douglas Laboratory of Richland, Wash. A longer-life power source in a 150-g model is planned.

BRITISH RADIOACTIVE-MATERIAL SALES*

The Radiochemical Centre's (Amersham) sales of radioactive materials reached £4.8 million ($13 million) for the fiscal year ending Mar. 31, 1971, about 60% of which represented exports. Since establishment of the Centre under public ownership in 1946, growth of sales has been rapid, averaging 15% per year in recent years, with consistently more than half the total exported. Currently, about 700 people are employed in the extensive laboratories and offices on an 18-acre site at Amersham and at a department in Harwell. The growth of sales was not limited to any particular products or markets, but particularly good progress was made in radioactive pharmaceuticals and radiation sources for industrial and medical use, and a substantial part of the sales was due to new products developed in recent years at Amersham. About 65% of a major new building complex—to provide additional laboratory and warehouse facilities—was commissioned and put in operation. A computerized order-processing system

*Excerpts from UKAEA annual report.

functioned satisfactorily, and the problems of decimalization were successfully overcome. A new comprehensive catalog was distributed worldwide to 25 thousand isotope users.

A planned increase in research and development effort resulted in further valuable developments in production methods and products, and important improvements were made in quality-control methods for additional reliability and high-purity products. Improvements in production of ^{14}C- and ^{3}H-labeled nucleosides and nucleotides and addition of 23 new ^{14}C- and 58 new ^{3}H-labeled compounds, many not available elsewhere, have put the Centre in a leading position as a supplier of these compounds. More than 90% of orders were met from stock.

The Amersham cyclotron operating systems have been developed to permit unattended running for both proton and deuteron irradiations, thereby providing extra irradiation time to meet the increasing demand for cyclotron-produced isotopes for medical and industrial use without increase in staff. Interlocks fitted to the operation systems—in some cases duplicated—provide for automatic shutdown of the machine in case of any deviation from normal conditions. An automatic data-logging system was designed to monitor the machine operation so that the reason for any trip-out can be ascertained.

The use of ^{125}I has grown so rapidly that it has become the most important isotope of iodine in the diagnostic nuclear medicine field. A new process to produce ^{125}I by irradiation of enriched ^{124}Xe under pressure in a reactor has given high yields of ^{125}I of 100% isotopic abundance and excellent quality for protein labeling. The United Kingdom is now self-sufficient for the supply of this important isotope.

Sterile generators were developed for the short-lived medical-diagnosis radionuclides ^{99m}Tc, ^{87m}Sr, and ^{113m}In. Not only can the products now be injected without a separate sterilization step, but also the product separation is easy and the product yield and concentration are high. These improvements have led to marked increases in sales in the United Kingdom and in Europe.

Curium-242 has been produced in considerable quantities by irradiation of ^{241}Am in the DIDO reactor. Two neutron sources containing a total of about a thousand curies of ^{242}Cm were produced with an output of 10^9 n/sec, which is exceptionally high for (α,n) sources. They are being used for reactor start-up—the first reported large-scale use of ^{242}Cm for this purpose.

INTERNATIONAL SYMPOSIUM ON RADIATION PRESERVATION OF FOOD

The IAEA and FAO will jointly sponsor a Symposium on the Radiation Preservation of Food in Bombay, India, Nov. 13–17, 1972. The meeting will cover recent advances in all aspects of food irradiation and will review and discuss the progress made since the 1966 Karlsruhe Symposium. The following list of topics has been scheduled:

I. Radiation sources and dosimetry.
II. Chemical and physical effects of radiation on food.
III. Wholesomeness of irradiated food.
IV. Physiological effects of radiation on foods of plant and animal origin.
V. Status of radiation preservation of various commodities:
  (*a*) Grain, stored food products, and animal feeds.
  (*b*) Fruits and vegetables.
  (*c*) Fish and seafoods.
  (*d*) Meat, meat products, and poultry.
  (*e*) Food ingredients (e.g., spices, texture aids, coloring matters, and enzymes).
  (*f*) Radiation-modified food products (resulting from modification of technological properties of foods and their constituents by irradiation).
VI. Facilities and economics of food irradiation.
VII. Legislation, control, and clearances of irradiated food.

For additional information, contact Miss C. de Mol van Otterloo, Division of External Relations, International Atomic Energy Agency, P. O. Box 590, Kärntner Ring 11, A-1011 Vienna, Austria.

EXECUTIVE DIRECTOR OF SINB

James T. Goodwin of Austin, Tex., has been named Executive Director of the Southern Interstate Nuclear Board (SINB), filling a vacancy created by the resignation of Robert H. Gifford to enter private industry. Goodwin joins the SINB from his position as Coordinator of Natural Resources in Texas, where he also was associated with various related projects.

CANRAD INDUSTRIES SUPPLIES TRITIUM-POWERED LIGHT SOURCES

Self-powered nuclear light sources with a useful life of 10 years or more, and independent of external power sources, consist of sealed glass tubes or bulbs, internally coated with a phosphor and filled with tritium gas. These Betalights,* which are manufactured

*Registered trademark.

by CANRAD Precision Industries of Pelham, N. Y., have typical brightnesses of 0.1 to 0.2 footlambert and are available in a variety of colors, depending on the phosphor used. They present no radiation hazard. Suggested uses are for dials and compasses, aircraft and military markers, photo darkroom markers, push buttons, and rifle-sight illuminators.

SYMPOSIUM ON NEW DEVELOPMENTS IN RADIOPHARMACEUTICALS AND LABELED COMPOUNDS

The IAEA will sponsor a Symposium in Copenhagen, Denmark, Mar. 26–30, 1973, which is designed to summarize the rapidly moving developments and related problems resulting from (1) the production and use of shorter-lived radioisotopes from "medical" cyclotrons; (2) the use of short-lived pharmaceuticals; and (3) the use of stable isotopes in medical research and diagnosis.

The program of the Symposium will emphasize the chemical and pharmaceutical aspects of the following topics:

I. Nuclide generators:
  (*a*) Problems associated with generator usage.
  (*b*) Improvements in generator design or chemistry.
  (*c*) New short-lived radioisotope generators.
II. Kits for preparing radiopharmaceuticals from short-lived isotopes.
III. Accelerator production of medically useful isotopes:
  (*a*) Improved equipment or methodology to facilitate routine use.
  (*b*) Targetry.
  (*c*) Isotope purification.
IV. Synthesis of radiopharmaceuticals and labeled compounds from short-lived isotopes.
V. Labeled compounds for *in vitro* clinical diagnostic procedures:
  (*a*) Radioimmunoassay.
  (*b*) Saturation analysis.
  (*c*) Detection of metabolic defects.
VI. Stable isotopes in medicine:
  (*a*) Improved isotopic enrichment procedures.
  (*b*) Synthesis of labeled compounds.
  (*c*) Medical applications.
VII. Regulatory and pharmaceutical responsibilities for radiopharmaceuticals:
  (*a*) International standards.
  (*b*) Regulation of radiopharmaceuticals at the research level and in routine use.
  (*c*) Pharmaceutical problems with generator- and accelerator-produced short-lived isotopes.
  (*d*) Regulation in developing countries.

Abstracts of papers must be submitted to the competent official authority of the Government of a Member State of the IAEA in time for transmission to and arrival at the Agency by Oct. 16, 1972. For further information, write Miss C. H. de Mol van Otterloo, Division of External Relations, International Atomic Energy Agency, P. O. Box 590, Kärntner Ring 11, A-1011 Vienna, Austria.

Abstracts of Recent Literature

Biological Effects of Radiation, J. E. Coggle and J. R. Noakes, St. Bartholomew's Hospital, London, and Uppingham School, Wykeham Science Series, Vol. 14, Wykeham Publications (London) Ltd., 1971 (Distributor: Springer-Verlag, New York) (148 pp., no references; $5.40).

Supplementary Keywords: *biology; physics; medicine; biochemistry; histology; effect of radiation on material; absorptiometry; secondary radiation effect; material tracing; dosimetry; safety; theory, applied; textbook.*

This book is mainly about the effects of ionizing radiation on mammals, especially man. The emphasis is on the links between the physicochemical and the biological effects. The intriguing nature of this work and its scientific and practical significances are evident in *Biological Effects of Radiation*—a book intended for advanced high school and college students in physics, biology, and chemistry.

The opening chapter describes the physics and chemistry of the direct and indirect actions of the various types of ionizing radiation on living matter. A chapter follows on the effect of radiation at the molecular and subcellular levels; the effects on proteins and nucleic acids are summarized, and brief discussions are included on radiation damage to cell components. Three chapters are devoted to radiation effects at the cellular level; among the topics included are cell survival and recovery and radiosensitivity of the cell at different phases of the cell cycle. A sixth chapter describes tissue-level radiation effects; the tissues considered include nervous-system, gastrointestinal, bone-

marrow, gonadal, and skin. The genetic effects of ionizing radiation are covered in a separate chapter; these include qualitative and quantitative aspects of radiation-induced chromosome aberrations, genetic mutations that result, and the genetic hazards of environmental radiation to human populations. A short chapter mentions the biological and chemical factors that influence the effect of radiation. A chapter is included on radiation and cancer, and another on radiation lifeshortening. Radiation hazards and the standards for radiation protection are the subjects of the final (11th) chapter.

The brief summaries within the chapters facilitate comprehension of the text; a glossary, subject index, and suggestions for additional reading are supplementary materials useful to the intended audience.

(HPR)

Industrial Radiology Techniques, R. Halmshaw, Ministry of Defense, Great Britain, Wykeham Technological Series, Vol. 3, Wykeham Publications (London) Ltd., 1971 (Distributor: Springer-Verlag, New York) (277 pp., no references; $10.80).

Supplementary Keywords: *source, X-ray; source, gamma; source, neutron; accelerator; industry; technique; apparatus; instrument; method; safety, container; theory, applied; absorptiometry; backscatter; secondary radiation effect; autoradiography; textbook; review, technical;* ^{60}Co; ^{192}Ir.

This is a book intended to introduce young scientists and engineers to industrial radiology, to help them choose the correct radiological technique for a particular use, and to ensure that they get the best possible results from a radiographic inspection. Thus the book is a broad, practical guide.

There are nine chapters, one on each of the following subjects: principles; equipment; radiographic technique; technique—specific cases; performance—sensitivity and limitation; film interpretation; fluoroscopy and image intensifiers; special methods of radiography; and safety and radiation protection. With a minimum of mathematics but very explicitly, the principles are given regarding the generation, absorption, and scattering of X and gamma rays, the radiographic process, and the effects of the radiation energy on radiographic films and screens. The discussion of equipment is very thorough; it includes radiation sources (tubes, accelerators, radioisotopes), their properties, and their containers; fixed and portable machines and the associated laboratory requirements; and films and screens and the facilities required to process, view, and evaluate them. The technique itself receives greatest emphasis, effort being made to explain clearly the interplay of all parameters that affect the sensitivity of the radiography. The sources of specifications, standards, and codes of good practice are tabulated, and information is given on the availability of reference radiographs. A broad discussion of technique is followed by a general stepwise procedure for producing radiographs and then by 19 varied examples of radiographic techniques appropriate to specific applications. Image-quality indicators receive emphasis in the discussion of the sensitivity limits of radiography, the factors that control them, and the methods of measuring sensitivity for flow detection—the most frequent industrial application of radiography. From film interpretation, the nature and significance of specimen defects are deduced; the detailed guide to this phase of radiography tells how to interpret radiographs of welds, castings, and assemblies and presents a summary list of acceptance standards that is useful to a worker in reporting the degree of acceptability or serviceability of a specimen. The special methods of radiography considered are X ray–paper, xero, flash, electron, color, neutron, micro, and auto, and the very special technique of tomoradiography. The book concludes with information very important to the novice industrial radiographer—safety and radiation protection. The illustrations are abundant and appropriate; they contribute significantly to the excellent coverage of the subject and enhance the clarity of a presentation that—even without them—is very understandable. (HPR)

Biological Aspects of Radiation Protection, Proceedings of the International Symposium, Kyoto, Japan, Oct. 13–15, 1969, T. Sugahara and O. Hug (Eds.), Kyoto University, Japan, and University of Munich, Germany, Igaku Shoin Ltd., Tokyo, 1971 (Distributor: Springer-Verlag, New York) (273 pp., $18.90).

Supplementary Keywords: *biology; physics; medicine; histology; biochemistry; effect of radiation on material; safety; material tracing.*

The two sources for estimation of the quantitative bases of radiation protection are (1) epidemiological data on the frequency of radiation injuries in man following high doses and (2) radiobiological evidence on the relation between the frequency of high-dose injury to the frequency of low-dose or low-dose-rate injury. The second of these was reviewed at the Symposium on Biological Aspects of Radiation Protection. Eighty-five participants represented 18 countries; 28 papers were presented in five sessions as follows:

Session	No. of papers
General Review of Biological Aspects of Radiation Protection	4
Hematological and Cytogenetic Effects of Radiation	6
Chemical Protection in Mammals	7
Fundamentals of Radiation Protection and Recovery	4
Comparative Individual Radiosensitivity and Its Cellular Explanation	7

This book comprises the proceedings of the Symposium, which was jointly supported by the International Association for Radiation Research and the International Atomic Energy Agency. The purposes of the Symposium were to have an Asian conference of radiation biologists to stimulate research and cooperation in an area where very few studies have been done on radiation effects relating to radiation protection and to stimulate contributions in radiobiology with a view to radiation protection, parallel to the biology of radiation therapy.

Although much new information was presented on radiation effects in mammals and man, it was concluded that the establishment of radiation biology for radiation protection has only just begun. The Symposium aroused many unanswered questions and suggested the possibility of developing new chemicals for radiation protection—the possibility of chemical treatment after irradiation with induction of the repair process by various means—thus removing the apparent dead end to chemical protection. The lively discussions, the numerous references cited in each article, and both a subject and an author index are included. (HPR)

Man and Atom. Building a New World Through Nuclear Technology, by G. T. Seaborg and W. R. Corliss, E. P. Dutton & Co., Inc., New York, 1971 (411 pp., $10.00).

Supplementary Keywords: *review, popular; isotope use; isotope user.*

Man and Atom is designed to close the gap of understanding between the general public and those engaged in nuclear affairs. It is the comprehensive story of the atom's progress and promise and of man's relation to the atom. The intelligent, although nontechnical, reader will find that the level of writing is not too sophisticated; as an aid, 54 illustrations and a glossary of terms are provided. These eminently qualified authors discuss their subject—one of deep concern to them—in three parts: atomic tools, applying the tools, and the atom and society. They have preferred to cover many topics (none in great detail) in the 12 chapters of the book and to illustrate the broad scope of the atom's influence with appended lists of applications of radioisotopes in medicine and of universities cooperating in Atomic Energy Commission programs. They do not offer rebuttals to specific arguments on the health and safety aspects of nuclear power, nor do they discuss arms-related subjects. For each chapter, they provide a list of suggested readings to assist those who wish to pursue topics further. They describe atomic tools to build a new world; for power and more power; and for labels, bond breakers, and explosives. They show how the tools are used to gain more food and water, to improve cities, to engineer our planet, and to explore the worlds above and below us. They show how the atom is a moving force in society, especially at the international level, and how it can be used to create new understandings. They conclude by reemphasizing the message of the nuclear age—the need to constructively merge those forces that flow from man and from the atom. (HPR)

Development of ^{131}I Production Techniques, by R. Constant, Belgian Report BLG-442 (in French; English translation ORNL-tr-2430 by Martha Gerrard).

This report discusses a number of methods for producing and recovering ^{131}I, with particular emphasis on patents. The original bibliography is supplemented and cross-referenced in the translation. (MG)

New Developments in the Techniques of Radiometric Titration, by J. Tölgyessy, *Isotopenpraxis*, 7: 208-217 (1971).

A review of the literature (61 references) in English.

Application of Nuclear Technology to Art Identification Problems, report for Feb. 1, 1970–Feb. 28, 1971, Carnegie-Mellon University, Mellon Institute, Pittsburgh, Pa. 15213.

This third annual report on work sponsored by the Division of Isotopes Development and the National Gallery of Art reports results of work on neutron activation analysis of paints for lead white. The results indicate that sources of lead white can be distinguished from one another if other pigments and linseed oil are

removed after the irradiation of the samples and if data are processed for probable trace element concentration distributions. Some synthetic pigments as well as some metal alloys and ceramics can be similarly distinguished. Also reported are developments of a low-level counting system for ^{14}C in linseed oil for use in dating and a study of X-ray diffraction of lead white. (MG)

Research and Development Studies in Environmental Pollution and in Reactor Cooling Systems, Nuclear Engineering, Part XXI, R. H. Moen (Ed.), Chemical Engineering Progress Symposium Series, Vol. 66, No. 104, 1970, American Institute of Chemical Engineers, New York.

The "Research and Development Studies in Environmental Pollution" section in this book (pp. 141-220) is of special interest to persons in the isotope field. An article by E. J. Croke and J. J. Roberts of Argonne National Laboratory on the Chicago air pollution system model gives basic information for use in pollution studies of all kinds.

Except for an article on the fusion torch for use in waste processing, all the other articles in this section are related to Division of Isotopes Development (DID) work. Bernard Manowitz et al. (Brookhaven National Laboratory) describe the isotope-ratio method for tracing atmospheric sulfur pollutants. M. Steinberg and M. Beller, also of Brookhaven, review their work on the use of ionizing radiation to synthesize ozone for water treatment, while D. S. Ballantine and L. A. Miller, of the DID Office in Washington, summarize the potential role of radiation in direct waste-water treatment. The destruction of organic substances in waste water by ionizing radiation is reported by D. M. J. Compton, formerly of Gulf General Atomic.

The papers appear to be printed exactly as submitted, with no editing or even retyping, which puts the complete responsibility for context on the individual authors. (MG)

Application of Nuclear Energy to Increasing Agricultural Productivity, Conference Proceedings, Santiago, Chile, Jan. 9–12, 1968 (CONF-680112) (in Spanish and English).

At a 1968 conference in Santiago, the proceedings of which have been recently distributed, papers were presented on water availability, mutations, animal and plant tracer studies, soil studies, and food processing. Most of the papers are concerned with radioisotopes. (MG)

Nuclear Instrument Landing System, Phase I Final Report, September 1970–June 1971, by R. A. Kaminskas, USAEC Report SAN-828-1, TRW Systems Group, Inc., Redondo Beach, Calif. 90278, October 1971.

The Nuclear Instrument Landing System, being developed under joint sponsorship of the Division of Isotopes Development and the Federal Aviation Agency, is to provide guidance and independent position checks for aircraft descending for a landing under zero visibility. The preliminary design, described here, uses four ^{60}Co sources which would be positioned on a runway as beacons to guide the incoming aircraft. (MG)

Californium-252 Progress, No. 9, October 1971.

The October 1971 issue of this periodical contains, besides news items concerning ^{252}Cf, articles on ^{252}Cf applications in therapy, neutron radiography, exploration of natural resources, process control, pollution control, and nuclear safeguards. Short features and a bibliography are included.

Basic Radiation Protection Criteria, Recommendations of the National Council on Radiation Protection and Measurements, NCRP Report No. 39, Jan. 15, 1971, NCRP Publications, P. O. Box 30175, Washington, D. C. 20014 ($2.00).

Specification of High Activity Gamma-Ray Sources, ICRU Report 18, Oct. 15, 1970, International Commission on Radiation Units and Measurements, 4201 Connecticut Avenue, NW, Washington, D. C. 20008 ($2.50).

Laboratory for Production of Radioisotopes, by A. M. Martinez, E. Parra, M. J. Lezama, C. G. Dominquez, A. Morales, and V. Ley-Koo (in Spanish), *Rev. Mex. Fis., Suppl. Reactor,* **19**: 585-597 (1970).

The CNEN Mexico City radioisotope production facility is described.

Recommendations for the Use of Radioisotopic Methods for Determining Physicochemical Properties of Soils (in Russian), Izdatel'stvo literatury po stroitel' stvu, Moscow, 1971 (49 pp., no references).

This handbook gives instructions for determining soil density with gamma-transmission and gamma-

scattering devices, moisture determination with fast-neutron sources, and direction and rate of flow of underground water with radioactive tracers. It also includes instrument specifications and methods for calculating results. (MG)

Measurement of the Water Equivalent of Snow and Soil Moisture by Airborne Gamma Surveying, R. M. Kogan, I. M. Nazarov, and Sh. D. Fridman, Chap. 10 in *Gamma Spectrometry of Natural Environments and Formations* (translated from Russian), Israel Program for Scientific Translations, Jerusalem, 1971.*

Supplementary Keywords: *source, gamma; hydrology; meteorology; geophysics; soil study; mobile; theory, applied; comparison; data handling; USSR, ^{40}K; U; Th.*

This book chapter describes the further application of the distortion of the gamma field of natural radioisotopes in soils and rocks to measure the water equivalent of snow and the moisture in topsoil. The chapter is introduced with a brief description of the factors that affect snow distribution over a territory. Then the detailed theory is presented on two types of methods for measuring the water equivalent of snow from the snow-cover distortion of the gamma field in the surface air. The direct methods, which require measurements at a single time only, include one that uses two monochromatic lines, emitted by some radionuclide in the soil, having different energies. Another direct method uses the flux of direct quanta of some monochromatic line together with the flux of scattered quanta of the same line. The other type method is based on the differences between measurements made at two different times and requires exact knowledge of flight altitude, air pressure, and air temperature. The radioisotopes considered in the work are ^{40}K, uranium, and thorium, which are compared with respect to the measured water-equivalent values. Theory indicates that the errors of the direct and difference methods should not exceed 10%.

The results of actual airborne snow surveys are presented. The measurements were made by the two versions of the direct method over different types of landscapes for water equivalents from 2 to 25 g/cm^2. For water-equivalent values of ~10 g/cm^2, the errors ranged from ~3 to 9%—a considerable improvement over the ~30% error that may sometimes occur with conventional methods that require the actual collection of snow samples.

Snow surveys by gamma-ray measurements made from the air were done over a total territory of 1 to 2 million km^2 for routes from 10 to 30 km long. The water equivalents ranged from 20 to 25 g/cm^2 and the errors of the measurements, from 0.5 to 1.0 g/cm^2. A snow map, derived from data taken in this manner, is presented for a 1-million-km^2 area west of the Urals in March 1967; the route spacing was 80 km. The map shows the flight routes and the lines of equal water equivalent.

Similarly, the detailed theory is presented for a measurement of soil and rock moisture, and one example of airborne measurements is given. (HPR)

Development of Nuclear Analytical Techniques for Oil-Slick Identification. Phase IIA. Final Report, by H. R. Lukens, D. Bryan, N. A. Hialt, and H. L. Schlesinger, USAEC Report GULF-RT-A-10684, Gulf Radiation Technology, June 11, 1971.

Environmental Monitoring in Idaho for Metallic Poisons Using the Materials Testing Reactor, R. F. Makens (Ed.), Special Issue No. 2 of *J. Idaho Acad. Sci.*, August 1971.

These proceedings of a symposium at the University of Idaho, Apr. 16, 1971, consist of papers on mercury determination in human foods, pheasants, fish, and hair. The Materials Testing Reactor was reactivated for use in the activation analysis carried out. (MG)

The Use of Radioisotopically Labeled Analytical Reagents in Organic Chemistry, by G. Ayrey, D. Barnard, and T. H. Houseman, *Chem. Rev.*, **71**: 371-393 (1971).

The major applications of labeled reagents in organic chemistry appearing in the literature up until the end of 1969 are reviewed, with extensive documentation. Neither actual syntheses of labeled reagents and compounds nor methods for measuring radiation are discussed, but review articles in the fields are referenced. (230 references) (MG)

*See also:

1. J. L. Smith and D. W. Willen, Radio–Snow Gages: A Review of the Literature, *Isotop. Radiat. Technol.*, 2(1): 41-49 (Fall 1964).

2. J. L. Smith, Instrumentation for Snow Gaging—Yesterday, Today, and Tomorrow, *Isotop. Radiat. Technol.*, 4(3): 227-237 (Spring 1967).

3. M. Gerrard (Sum.), Measurement of Snow-Water Reserves Using Natural Soil Radioactivity, *Isotop. Radiat. Technol.*, 5(3): 212-213 (Spring 1968).

Cobalt-60 Heat Source Demonstration Program. Phase II, Final Report. Detail Design, by J. W. Sadler, USAEC Report WANL-543-4, Westinghouse Electric Corporation Astronuclear Laboratory, May 1971.

Reaction Cell Improvements in the Mercury Substitution–Nuclear Detection Sulfur Dioxide Stack Gas Monitor, Final Report, Mar. 18, 1970–Oct. 14, 1971, by J. E. Howes, Jr., H. S. Rosenberg, and R. A. Ewing, USAEC Report TID-25850, Battelle Columbus Laboratories, Oct. 14, 1971.

Mineral Studies with Isotopes in Domestic Animals, Panel Proceedings, Vienna, Sept. 28–Oct. 2, 1970, IAEA, Vienna, 1971 (STI/PUB/293) (203 pp., $6.00).

Because the production of animal protein is greatly affected by deficiency or oversupply of minerals to livestock, the proceedings of this panel should be useful to persons working on increasing the world's protein supply. It was attended by 16 representatives of 13 countries, who are experienced in the use of nuclear techniques for studying mineral metabolism in domestic animals. Papers were presented on the commonly recognized trace elements—including Zn, I, Ca, P, Co, Cu, Fe, and Mn—which were studied with radioisotopic tracers. An extensive review of the literature on zinc metabolism has 180 references. Numerous original studies are presented or discussed with descriptions of the techniques used in administering the radioisotopes to cattle, rabbits, poultry, swine, sheep, goats, and buffaloes as well as descriptions of such unwieldy apparatus as a whole body counter for cattle and equipment for restraining the animals during the experiments. Most of the papers presented are well documented. The panel was organized by the Joint FAO/IAEA Division of Atomic Energy in Food and Agriculture and was the second of its kind. The first was held in 1968.* (MG)

**Trace Mineral Studies with Isotopes in Domestic Animals,* Panel Proceedings, Vienna, Oct. 28–31, 1968, IAEA, Vienna, 1969 (STI/PUB/218).

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